Spin-orbit crossed susceptibility in topological Dirac semimetals

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We theoretically study the spin-orbit crossed susceptibility of topological Dirac semimetals. Because of strong spin-orbit coupling, the orbital magnetization is induced by Zeeman coupling. We find that the spin-orbit crossed susceptibility is proportional to the separation of the Dirac points and it is highly anisotropic. The orbital magnetization is induced only along the rotational symmetry axis. We also study the conventional spin susceptibility. The spin susceptibility exhibits anisotropy and the spin magnetization is induced only along the perpendicular to the rotational symmetry axis in contrast to the spin-orbit crossed susceptibility. We quantitatively compare the two susceptibilities and find that they can be comparable.

I. INTRODUCTION

In the presence of an external magnetic field, magnetization is induced by both the orbital motion and spin magnetic moment of electrons. The magnetization is composed of the orbital and spin magnetization, which are induced by the minimal substitution, \( p \rightarrow p + eA \), and the Zeeman coupling, respectively. Additionally, spin-orbit coupling gives rise to the spin-orbit crossed response, in which the spin magnetization is induced by the minimal substitution, and the orbital magnetization is induced by the Zeeman coupling. In the strongly spin-orbit coupled systems, the spin-orbit crossed response can give comparable contribution to the conventional spin and orbital magnetic responses. On the other hand, the spin-orbit coupling plays a key role to realize a topological phase of matter, for example topological insulators and topological semimetals. A natural question arising is what kind of the spin-orbit crossed response occurs in the topological materials. Because of the topologically nontrivial electronic structure and the existence of the topological surface states, the topological materials exhibit the spin-orbit crossed response as a topological response.

The topological Dirac semimetal is one of the topological semimetals and experimentally observed in Na₃Bi and Cd₃As₂. The topological Dirac semimetal has an inverted band structure originating from strong spin-orbit coupling and the Dirac points are protected by rotational symmetry. A remarkable feature of the topological Dirac semimetal is the conservation of the spin angular momentum along the rotation axis within a low energy approximation. The topological Dirac semimetal is regarded as layers of two-dimensional (2D) quantum spin Hall insulators (QSHI) staked in momentum space, and exhibits the intrinsic semi-quantized spin Hall effect.

The magnetic responses of the generic Dirac electrons have been investigated in several theoretical papers. The orbital susceptibility logarithmically diverges and exhibits strong diamagnetism at the Dirac point. When spin-orbit coupling is not negligible, the spin susceptibility becomes finite even at the Dirac point where the density of states vanishes. This is contrast to the conventional Pauli paramagnetism and known as the Van Vleck paramagnetism.

In this paper, we study the spin-orbit crossed susceptibility of the topological Dirac semimetal. We find that the spin-orbit crossed susceptibility is proportional to the separation of the Dirac points and independent of the microscopic parameters of the materials. We also include the spin conservation breaking term which mixes up and down spins. We confirm that the spin-orbit crossed susceptibility is approximately proportional to the separation of the Dirac points even in the absence of the spin conservation as long as the separation is small. We also calculate the spin susceptibility and quantitatively compare the two susceptibilities. Using the material parameters for Na₃Bi and Cd₃As₂, we show that the spin-orbit crossed susceptibility can give quite large contribution to the total magnetic susceptibility.

The paper is organized as follows. In Sec. II, we introduce a model Hamiltonian. In Secs. III and IV, we calculate the spin-orbit crossed susceptibility and the spin susceptibility. In Secs. V and VI, the discussion and conclusion are given.

II. MODEL HAMILTONIAN

We consider a model Hamiltonian on the cubic lattice

\[
H_k = H_{\mathrm{TDS}} + H_{\mathrm{xy}} + H_{\mathrm{Zeeman}},
\]

which is composed of three terms. The first and second terms describe the electronic states in the topological Dirac semimetals, which reduces to the low energy effective Hamiltonian around the Γ point. The first term is given by

\[
H_{\mathrm{TDS}} = \varepsilon_k + \tau_x \sigma_z t \sin(k_x a) - \tau_y t \sin(k_y a) + \tau_z m_k,
\]

where

\[
\varepsilon_k = C_0 - C_1 \cos(k_z c) - C_2 [\cos(k_x a) + \cos(k_y a)],
\]

\[
m_k = m_0 + m_1 \cos(k_z c) + m_2 [\cos(k_x a) + \cos(k_y a)].
\]
and $a$ and $c$ are the lattice constants. Pauli matrices $\sigma$ and $\tau$ act on real and pseudo spin (orbital) degrees of freedom. There are Dirac points at $(0,0,\pm k_D)$,

$$k_D = \frac{1}{c} \arccos \left( -\frac{m_0 + 2m_n}{m_1} \right).$$

The first term, $H_{\text{TDs}}$, commutes with the spin operator $\sigma_z$, i.e. the $z$-component of spin conserves, and the Hamiltonian is regarded as the Bernevig-Hughes-Zhang model \[20\] extended to three-dimension. The second term is given by

$$H_{\text{xy}} = \tau_x \sigma_x \gamma [\cos(k_x a) - \cos(k_y a)] \sin(k_x c) + \tau_x \sigma_y \gamma \sin(k_y a) \sin(k_x c),$$

where $[23-27]$, which mixes up and down spins. When $H_{\text{xy}}$ is expanded around the $\Gamma$ point, leading order terms are third order terms, which are related to three-fold rotational symmetry around the $\Gamma$ point, leading order terms are third order terms and its eigenenergy is $2\gamma$.

III. SPIN-ORBIT CROSSED SUSCEPTIBILITY

A. Formulation

The orbital magnetization is calculated by the formula \[3, 4, 28\],

$$\chi_{\alpha\beta} = \frac{\partial M_{\alpha}^{\text{orbit}}}{\partial \mu},$$

$$\sigma_{\alpha\beta} = -e\varepsilon_{\alpha\beta} \left( \frac{\partial M_{\alpha}^{\text{orbit}}}{\partial \mu} \right),$$

where $M_{\alpha}^{\text{orbit}}$ is the orbital magnetization operator and given by

$$M_{\alpha}^{\text{orbital}} = \frac{\mu_B}{2} \left( g_s \sigma_\alpha 0 \right) \left( 0 g_p \sigma_\alpha \right) = g_+ \mu_B \tau_0 \sigma + g_- \mu_B \tau_z \sigma.$$

The spin-orbit crossed susceptibility is derived by \[5, 6\],

$$\chi_{\alpha\beta} = \frac{\partial M_{\alpha}^{\text{orbit}}}{\partial B_{\beta}} = \frac{\partial M_{\alpha}^{\text{spin}}}{\partial B_{\beta}}.$$
points $k_D$. In the present model, there are several parameters, such as $t, a, m_0$, and so on. We systematically change them and find which parameter affect the value of $\chi_{zz}^{SO}$. Figure 2(a), (b), and (c) show that $\chi_{zz}^{SO}$ increases linearly with $k_D$ and satisfy following relation,

$$\chi_{zz}^{SO} = g + \mu_B \frac{2e}{k} \frac{k_D}{\pi},$$

(14)

$\chi_{zz}^{SO}$ is proportional to the separation of the Dirac points $k_D$ and the coupling constant $g + \mu_B$. This result is understood as follows. $\chi_{zz}^{SO}$ is obtained as

$$\chi_{zz}^{SO} = \int_{-\pi/c}^{\pi/c} \frac{dk_z}{2\pi} \chi_{zz}^{SO(2D)}(k_z),$$

(15)

where $\chi_{zz}^{SO(2D)}$ is the 2D spin-orbit crossed susceptibility at fixed $k_z$. $\chi_{zz}^{SO(2D)}$ is quantized as $2g + \mu_B e/\hbar$ in the 2D quantum spin Hall insulators and vanishes in the ordinary insulators [4, 6]. The topological Dirac semimetal is regarded as layers of the 2D-QSHI thickened in the momentum space and the spin Chern number on the momentum space and the spin Chern number on the

In the presence of $H_{xy}$, $\gamma = 0$, $\chi_{zz}^{SO}$ is finite as we mentioned above. On the other hand, $\chi_{xx}^{SO}$ and $\chi_{yy}^{SO}$ are zero. This means that the orbital magnetization is induced only along $z$-axis, which is the rotational symmetry axis. At finite $\gamma$, $\chi_{zz}^{SO}$ is an even function and $\chi_{z\gamma(y\gamma)}^{SO}$ is an odd function.

Figure 2(a) shows $\chi_{zz}^{SO}$ around the Dirac point as a function of $\varepsilon_F$. When $g_-/g_+ = 0$, $\chi_{zz}^{SO}$ is an even function around the Dirac point. At $\varepsilon_F = 0$, $\chi_{zz}^{SO}$ is independent of $g_-/g_+$ as we see it in Fig. 2(b). When $g_-/g_+ \neq 0$, however, $\chi_{zz}^{SO}$ is asymmetric and the derivative of $\chi_{zz}^{SO}$ is finite. The sign of the derivative corresponds to the sign of $g_+/g_-$. This suggests that the Hall conductivity is finite when $g_-/g_+ \neq 0$. Calculating Eq. (12), We confirm that the Hall conductivity is finite at $\varepsilon_F = 0$. Figure 2(b) shows $\sigma_{xy}$ as a function of $g_+/g_-$. $\sigma_{xy}$ linearly increases with $g_+/g_-$. The topological Dirac semimetal is viewed as a time reversal pair of the Weyl semimetal with up/down spin. Therefore, the Hall conductivity completely cancel with each other. Even in the presence of $g_-$ Zeeman coupling (the orbital independent term), the cancellation is retained. In the presence of $g_-$ Zeeman term (the orbital dependent term), on the other hand, the cancellation is broken. This is because $g_-$ Zeeman coupling changes the separation of the Dirac points opposite direction for the up and down spin Weyl semimetals. As a result, the Hall conductivity is finite in $g_-/g_+ \neq 0$ and given by

$$\sigma_{xy} = \frac{2e^2}{\pi \hbar a} \frac{g_- \mu_B B_{\text{spin}}}{t}.$$  

(16)

This expression is quantitatively consistent with the numerical result in Fig. 2(b).
where \( \chi \) is the long wavelength limit of the distribution function, where \( \varepsilon \) is the energy of \( k \). FIG. 3: The spin-orbit crossed susceptibility \( \chi \) as a function of \( \varepsilon \). We set the parameters \( m_0 = -2m, m_1 = m, m_1/t = 1, c/a = 1, \) and \( \gamma = 0 \).

IV. SPIN SUSCEPTIBILITY

In this section, we calculate the spin susceptibility using the Kubo formula,

\[
\chi_{\alpha\alpha}(q, \varepsilon_F) = \frac{1}{V} \sum_{nk} \frac{-f_{nk} + f_{m-k-q}}{\varepsilon_{nk} - \varepsilon_{m-k-q}} \times |\langle n, k | M_{\alpha}^{\text{spin}} | m, k - q \rangle|^2,
\]

where \( V \) is the volume of the system, \( f_{nk} \) is the Fermi distribution function, \( \varepsilon_{nk} \) is energy of \( n \)-th band and \( |n, k \rangle \) is a Bloch state of the unperturbed Hamiltonian. Taking the long wavelength limit \( |q| \to 0 \), we obtain

\[
\lim_{|q| \to 0} \chi_{\alpha\alpha}^{\text{spin}}(q, \varepsilon_F) = \chi_{\alpha\alpha}^{\text{intra}}(\varepsilon_F) + \chi_{\alpha\alpha}^{\text{inter}}(\varepsilon_F),
\]

(17)

where \( \chi_{\alpha\alpha}^{\text{intra}}(\varepsilon_F) \) is an intraband contribution,

\[
\chi_{\alpha\alpha}^{\text{intra}}(\varepsilon_F) = \frac{1}{V} \sum_{nk} \left( -\frac{\partial f_{nk}}{\partial \varepsilon_{nk}} \right) |\langle n, k | M_{\alpha}^{\text{spin}} | m, k \rangle|^2,
\]

(18)

and \( \chi_{\alpha\alpha}^{\text{inter}}(\varepsilon_F) \) is an interband contribution,

\[
\chi_{\alpha\alpha}^{\text{inter}}(\varepsilon_F) = \frac{1}{V} \sum_{nk \neq m, k} \frac{-f_{nk} + f_{m-k}}{\varepsilon_{nk} - \varepsilon_{m-k}} \times |\langle n, k | M_{\alpha}^{\text{spin}} | m, k \rangle|^2.
\]

(19)

At the zero temperature, only electronic states on the Fermi surface contribute to \( \chi_{\alpha\alpha}^{\text{intra}} \). On the other hand, all electronic states below the Fermi energy can contribute to \( \chi_{\alpha\alpha}^{\text{inter}} \). From the above expression, we see that \( \chi_{\alpha\alpha}^{\text{inter}} \) becomes finite, when the matrix elements of the spin operator between the conduction and valence bands are non-zero, i.e. the commutation relation between the Hamiltonian and the spin operator is non-zero. If the Hamiltonian and the spin operator commute,

\[
\langle n, k | [H_k, M_{\alpha}^{\text{spin}}] | m, k \rangle = 0,
\]

(21)

the interband matrix element satisfies

\[
(\varepsilon_{nk} - \varepsilon_{mk}) \langle n, k | M_{\alpha}^{\text{spin}} | m, k \rangle = 0.
\]

(22)

This equation means that there is no interband matrix element and \( \chi_{\alpha\alpha}^{\text{inter}} = 0 \), because \( \varepsilon_{nk} - \varepsilon_{mk} \neq 0 \).

Figure 4 shows the spin susceptibility \( \chi_{\alpha\alpha}^{\text{spin}} \) at \( \varepsilon_F = 0 \) as a function of (a) \( \gamma \) and (b) \( g_-/g_+ \). At \( \varepsilon_F = 0 \), the Density of states vanishes, so that \( \chi_{\alpha\alpha}^{\text{intra}} \) gives no contribution and \( \chi_{\alpha\alpha}^{\text{spin}} \) is solely given by \( \chi_{\alpha\alpha}^{\text{inter}} \). In the following, we explain the qualitative behavior of \( \chi_{\alpha\alpha}^{\text{SO}} \) using the commutation relation between the Hamiltonian and spin operator. In Fig. 4 (a), \( \chi_{\alpha\alpha}^{\text{spin}} \) vanishes at \( \gamma = 0 \), because the Hamiltonian, \( H_{\text{TDS}} \), and the spin operator of \( z \)-component, \( g_+ B_0 \tau_z \), commute,

\[
[H_{\text{TDS}}, g_+ \mu_B \tau_z \sigma_z] = 0.
\]

(23)

Away from \( \gamma = 0 \), on the other hand, \( \chi_{\alpha\alpha}^{\text{SO}} \) increases with \( |\gamma| \). This is because the commutation relation between \( H_{\text{xy}} \) and \( g_+ B_0 \tau_\sigma \) is non-zero,

\[
[H_{\text{xy}}, g_+ \mu_B \tau_\sigma \sigma_z] = 0,
\]

(24)

and \( \chi_{\alpha\alpha}^{\text{inter}} \) gives finite contribution. \( \chi_{\alpha\alpha}^{\text{spin}} \) and \( \chi_{\alpha\alpha}^{\text{spin}} \) are finite even in the absence of \( H_{\text{xy}} \), i.e. \( \gamma = 0 \), because \( H_{\text{TDS}} \) and \( g_+ \mu_B \tau_\sigma \) do not commute,

\[
[H_{\text{TDS}}, g_+ \mu_B \tau_\sigma \sigma_x] = 0,
\]

(25)

At \( \gamma = 0 \), \( \chi_{\alpha\alpha}^{\text{spin}} \) is equal to \( \chi_{\alpha\alpha}^{\text{spin}} \). Away from \( \gamma = 0 \), however, they deviate from each other. This is because \( H_{\text{TDS}} \) possesses four-fold rotational symmetry along \( z \)-axis but \( H_{\text{xy}} \) breaks the four-fold rotational symmetry. Figure 4 (b) shows that \( \chi_{\alpha\alpha}^{\text{SO}} \) becomes finite when \( g_-/g_+ \neq 0 \). The orbital dependent term, \( g_- \mu_B \tau_z \sigma_z \), and \( H_{\text{TDS}} \) do not commute,

\[
[H_{\text{TDS}}, g_- \mu_B \tau_z \sigma_z] = 0.
\]

(26)

Consequently, \( \chi_{\alpha\alpha}^{\text{inter}} \) gives finite contribution, though the \( z \)-component of spin is a good quantum number. The orbital dependent term does not break the four-fold rotational symmetry along \( z \)-axis, so that \( \chi_{\alpha\alpha}^{\text{spin}} \) is equal to \( \chi_{\alpha\alpha}^{\text{spin}} \) in Fig. 4 (b).

The spin susceptibility \( \chi_{\alpha\alpha}^{\text{spin}} \) is also anisotropic but contrasts with the spin-orbit crossed susceptibility \( \chi_{\alpha\alpha}^{\text{SO}} \). \( \chi_{\alpha\alpha}^{\text{spin}} \) and \( \chi_{\alpha\alpha}^{\text{spin}} \) are larger than \( \chi_{\alpha\alpha}^{\text{spin}} \), in contrast \( \chi_{\alpha\alpha}^{\text{SO}} \) is larger than \( \chi_{\alpha\alpha}^{\text{SO}} \) and \( \chi_{\alpha\alpha}^{\text{SO}} \). Therefore, the angle dependence measurement of magnetization will be useful to separate the contribution from the each susceptibility.
In the present parameters, $\chi^{\text{SO}}_{zz}$ is negative and depends on $g_{-}/g_{+}$. The dependence on $g_{-}/g_{+}$ originates from the existence of $\varepsilon_{k}$, which breaks the particle-hole symmetry. The $g$-factors are experimentally estimated as $g_{z} = 18.6$ for Cd$_{2}$As$_{3}$ \cite{30} and $g_{-} = 20$ for Na$_{3}$Bi \cite{7,11}. Unfortunately, there is no experimental data which determines both of $g_{s}, g_{p}$ or $g_{-}, g_{+}$. From Fig. 5 we see that $\chi^{\text{SO}}_{zz}$ is dominant if $g_{-}/g_{+} \geq 0$.}

| Material parameters | Cd$_{2}$As$_{3}$ | Na$_{3}$Bi |
|---------------------|------------------|------------|
| $C_{0}$            | 0.306[eV]        | -1.183[eV]|
| $C_{1}$            | 0.033[eV]        | 0.188[eV]  |
| $C_{2}$            | 0.144[eV]        | -0.654[eV]|
| $m_{0}$            | 0.376[eV]        | 1.754[eV]  |
| $m_{1}$            | -0.058[eV]       | -0.228[eV]|
| $m_{2}$            | -0.169[eV]       | -0.806[eV]|
| $t$                | 0.070[eV]        | 0.485[eV]  |
| $A$                | 12.64[A]         | 5.07[A]    |
| $C$                | 25.43[A]         | 9.66[A]    |

VI. CONCLUSION

We theoretically study the spin-orbit crossed susceptibility of topological Dirac semimetals. We find that the spin-orbit crossed susceptibility along rotational symmetry axis is proportional to the separation of the Dirac points and is independent of the microscopic model parameters. The spin-orbit crossed susceptibility is induced only along the rotational symmetry axis. We also cal-
ulate the spin susceptibility. The spin susceptibility is anisotropic and vanishingly small along the rotational symmetry axis, in contrast to the spin-orbit crossed susceptibility. The two susceptibilities are quantitatively compared for material parameters of Cd$_2$As$_3$ and Na$_3$Bi. The spin-orbit crossed susceptibility can be dominant contribution for the total susceptibility.

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