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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 motion of electrons is modulated by Zeeman coupling, which contributes to orbital magnetization. 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. When spin-orbit coupling is negligible, the magnetization is composed of the orbital and spin magnetization, which are induced by the minimal substitution, $\mathbf{p} \rightarrow \mathbf{p} + e\mathbf{A}$, 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.

Spin-orbit coupling plays a key role to realize a topological phase of matter, such as topological insulators [1] and topological semimetals [2]. 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 [3–7]. The spin-orbit crossed response has been investigated in several systems. In the literature the connection between the spin-orbit crossed susceptibility and the spin Hall conductivity was pointed out [3, 4]. In recent theoretical work, the spin-orbit crossed response has been investigated also in Rashba spin-orbit coupled systems [8, 9].

The topological Dirac semimetal is one of the topological semimetals [10–15] and experimentally observed in Na_3Bi and Cd_3As_2 [16–18]. The topological Dirac semimetals have an inverted band structure originating from strong spin-orbit coupling. They are characterized by a pair of Dirac points in the bulk and Fermi arcs on the surface [10, 11]. The Dirac points are protected by rotational symmetry along the axis perpendicular to the (001) surface in the case of Na_3Bi and Cd_3As_2 [10, 11]. This is an important difference from the Dirac semimetals appearing at the phase boundary of topological insulators and ordinary insulators [19–22], in which there

is no Fermi arc. A remarkable feature of the topological Dirac semimetals is the conservation of the spin angular momentum along the rotation axis within a low energy approximation [23]. The topological Dirac semimetals are regarded as layers of two-dimensional (2D) quantum spin Hall insulators (QSHI) stacked in momentum space and exhibit 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 [6, 24–26]. When spin-orbit coupling is not negligible, the spin susceptibility becomes finite even at the Dirac point where the density of states vanishes [6, 27–29]. This is contrast to the conventional Pauli paramagnetism and known as the Van Vleck paramagnetism [29–32].

In this paper, we study the spin-orbit crossed susceptibility of the topological Dirac semimetals. We find that the spin-orbit crossed susceptibility is proportional to the separation of the Dirac points and independent of the other microscopic parameters of the materials. We also include the spin conservation breaking term which mixes up and down spins [10, 11]. 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 sufficiently small. We also calculate the spin susceptibility and quantitatively compare the two susceptibilities. Using the material parameters for Na_3Bi and Cd_3As_2 , we show that the contribution of the spin-orbit crossed susceptibility is important in order to appropriately estimate the total susceptibility.

The paper is organized as follows. In Sec. II, we introduce a model Hamiltonian and define the spin-orbit crossed susceptibility. 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

$$H_{\mathbf{k}} = H_{\text{TDS}} + H_{\text{xy}} + H_{\text{Zeeman}}, \quad (1)$$

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 [10–12, 14, 15]. The first term is given by

$$H_{\text{TDS}} = \varepsilon_{\mathbf{k}} + \tau_x \sigma_z t \sin(k_x a) - \tau_y t \sin(k_y a) + \tau_z m_{\mathbf{k}}, \quad (2)$$

where

$$\begin{aligned} \varepsilon_{\mathbf{k}} &= C_0 - C_1 \cos(k_z c) - C_2 [\cos(k_x a) + \cos(k_y a)], \\ m_{\mathbf{k}} &= m_0 + m_1 \cos(k_z c) + m_2 [\cos(k_x a) + \cos(k_y a)]. \end{aligned} \quad (3)$$

Pauli matrices $\boldsymbol{\sigma}$ and $\boldsymbol{\tau}$ act on real and pseudo spin (orbital) degrees of freedom. a and c are the lattice constants. t , C_1 , and C_2 are hopping parameters. C_0 gives constant energy shift. m_0 , m_1 , and m_2 are related to strength of spin-orbit coupling and lead band inversion. There are Dirac points at $(0, 0, \pm k_D)$,

$$k_D = \frac{1}{c} \arccos \left(-\frac{m_0 + 2m_2}{m_1} \right). \quad (4)$$

The separation of the Dirac points is tuned by changing the parameters, m_0 , m_1 , and m_2 . The first term, H_{TDS} , commutes with the spin operator σ_z , and H_{TDS} is regarded as the Bernevig-Hughes-Zhang model [12, 33] extended to three-dimension. The second term is given by

$$\begin{aligned} H_{\text{xy}} &= \tau_x \sigma_x \gamma [\cos(k_y a) - \cos(k_x a)] \sin(k_z c) \\ &\quad + \tau_x \sigma_y \gamma \sin(k_x a) \sin(k_y a) \sin(k_z c), \end{aligned} \quad (5)$$

which mixes up and down spins. When H_{xy} is expanded around the Γ point, leading order terms are third order terms, which are related to the rotational symmetry along the axis perpendicular to the (001) surface in Na_3Bi and Cd_3As_2 . In the current system, this axis corresponds to the z -axis and we call it the rotational symmetry axis in the following. γ corresponds to the coefficient of the third order terms in the effective model [10, 11]. When γ is zero, the z -component of spin conserves. At finite γ , on the other hand, the z -component of spin is not conserved.

As we mentioned in the introduction, the external magnetic field enters the Hamiltonian via the minimal substitution, $\mathbf{p} \rightarrow \mathbf{p} + e\mathbf{A}$, and the Zeeman coupling. We formally distinguish the magnetic field by the way it enters the Hamiltonian in order to extract the spin-orbit crossed response. $\mathbf{B}^{\text{orbit}}$ and \mathbf{B}^{spin} represent the magnetic field in the minimal substitution and in the Zeeman coupling respectively. They are the same quantities so that we have to set $\mathbf{B}^{\text{orbit}} = \mathbf{B}^{\text{spin}}$ at the end of the calculation.

In the following, the subscripts $\alpha, \beta, \gamma, \delta$ refer to x, y, z . We define the orbital magnetization $M_{\alpha}^{\text{orbit}}$ and the spin magnetization M_{α}^{spin} as follows

$$M_{\alpha}^{\text{orbit}} = -\frac{1}{V} \frac{\partial \Omega}{\partial B_{\alpha}^{\text{orbit}}}, \quad (6)$$

$$M_{\alpha}^{\text{spin}} = -\frac{1}{V} \frac{\partial \Omega}{\partial B_{\alpha}^{\text{spin}}}, \quad (7)$$

where Ω is the thermodynamic potential and V is the system volume. These quantities are written, up to linear order in $\mathbf{B}^{\text{orbit}}$ and \mathbf{B}^{spin} , as

$$M_{\alpha}^{\text{orbit}} = \chi_{\alpha\beta}^{\text{orbit}} B_{\beta}^{\text{orbit}} + \chi_{\alpha\beta}^{\text{SO}} B_{\beta}^{\text{spin}}, \quad (8)$$

$$M_{\alpha}^{\text{spin}} = \chi_{\alpha\beta}^{\text{spin}} B_{\beta}^{\text{spin}} + \chi_{\alpha\beta}^{\text{SO}} B_{\beta}^{\text{orbit}}, \quad (9)$$

where

$$\chi_{\alpha\beta}^{\text{orbit}} = \frac{\partial M_{\alpha}^{\text{orbit}}}{\partial B_{\beta}^{\text{orbit}}}, \quad (10)$$

$$\chi_{\alpha\beta}^{\text{spin}} = \frac{\partial M_{\alpha}^{\text{spin}}}{\partial B_{\beta}^{\text{spin}}}, \quad (11)$$

$$\chi_{\alpha\beta}^{\text{SO}} = \frac{\partial M_{\alpha}^{\text{orbit}}}{\partial B_{\beta}^{\text{spin}}} = \frac{\partial M_{\alpha}^{\text{spin}}}{\partial B_{\beta}^{\text{orbit}}}. \quad (12)$$

Spin-orbit coupling can give the spin-orbit crossed susceptibility $\chi_{\alpha\beta}^{\text{SO}}$, in addition to the conventional spin and orbital susceptibilities, $\chi_{\alpha\beta}^{\text{spin}}$ and $\chi_{\alpha\beta}^{\text{orbit}}$ [6, 7].

In the rest of the paper, we focus on the Zeeman coupling, which can induce both of the orbital and spin magnetization as we see in Eqs. (8) and (9). The Zeeman coupling is given by

$$\begin{aligned} H_{\text{Zeeman}} &= -\frac{\mu_B}{2} \begin{pmatrix} g_s \boldsymbol{\sigma} & 0 \\ 0 & g_p \boldsymbol{\sigma} \end{pmatrix} \cdot \mathbf{B}^{\text{spin}}, \\ &= -g_+ \mu_B \tau_0 \boldsymbol{\sigma} \cdot \mathbf{B}^{\text{spin}} - g_- \mu_B \tau_z \boldsymbol{\sigma} \cdot \mathbf{B}^{\text{spin}}, \end{aligned} \quad (13)$$

where μ_B is the Bohr magneton and g_s, g_p correspond to the g -factors of electrons in s and p orbitals, respectively. We define $g_+ = (g_s + g_p)/4$ and $g_- = (g_s - g_p)/4$, so that the Zeeman coupling contains two terms, the symmetric term $\tau_0 \boldsymbol{\sigma}$ and the antisymmetric term $\tau_z \boldsymbol{\sigma}$ [7, 34, 35].

III. SPIN-ORBIT CROSSED SUSCEPTIBILITY

A. Formulation

The orbital magnetization is calculated by the formula [36–40],

$$\begin{aligned} M_{\alpha}^{\text{orbit}} &= \frac{e}{2\hbar} \sum_n \int_{\text{BZ}} \frac{d^3 k}{(2\pi)^3} f_{n\mathbf{k}} \epsilon_{\alpha\beta\gamma} \\ &\quad \times \text{Im} \langle \partial_{\beta} n, \mathbf{k} | (\varepsilon_{n\mathbf{k}} + H_{\mathbf{k}} - 2\mu) | \partial_{\gamma} n, \mathbf{k} \rangle, \end{aligned} \quad (14)$$

where $f_{n\mathbf{k}} = [1 + e^{(\varepsilon_{n\mathbf{k}} - \mu)/k_B T}]^{-1}$ is the Fermi distribution function, $\partial_{\alpha} = \frac{\partial}{\partial k_{\alpha}}$, and $|n, \mathbf{k}\rangle$ is a eigenstate of $H_{\mathbf{k}}$

and its eigenenergy is $\varepsilon_{n\mathbf{k}}$. The derivative of the eigenstates $|\partial_\alpha n, \mathbf{k}\rangle$ is expanded as [39]

$$|\partial_\alpha n, \mathbf{k}\rangle = c_n |n, \mathbf{k}\rangle + \sum_{m \neq n} \frac{\langle m, \mathbf{k} | \hbar v_\alpha | n, \mathbf{k} \rangle}{\varepsilon_{m\mathbf{k}} - \varepsilon_{n\mathbf{k}}} |m, \mathbf{k}\rangle, \quad (15)$$

where the velocity operator v_α is given by $v_\alpha = \partial_\alpha H_{\mathbf{k}}/\hbar$ and c_n is a pure imaginary number. Using Eq. (15), the formula, Eq. (14), is written as

$$M_\alpha^{\text{orbit}} = \frac{e}{2\hbar} \sum_n \int_{\text{BZ}} \frac{d^3k}{(2\pi)^3} f_{n\mathbf{k}} \epsilon_{\alpha\beta\gamma} \times \text{Im} \sum_{m \neq n} \frac{\langle n, \mathbf{k} | \hbar v_\beta | m, \mathbf{k} \rangle \langle m, \mathbf{k} | \hbar v_\gamma | n, \mathbf{k} \rangle}{(\varepsilon_{m\mathbf{k}} - \varepsilon_{n\mathbf{k}})^2} (\varepsilon_{n\mathbf{k}} + \varepsilon_{m\mathbf{k}} - 2\mu). \quad (16)$$

We use the above formula in numerical calculation. Using the 2D orbital magnetization $M_z^{\text{orbit}(2D)}(k_z)$ at fixed k_z , M_z^{orbit} is expressed as

$$M_z^{\text{orbit}} = \int_{-\pi/c}^{\pi/c} \frac{dk_z}{2\pi} M_z^{\text{orbit}(2D)}(k_z). \quad (17)$$

The above expression is useful when we discuss numerical results for χ_{zz}^{SO} . We can relate $\chi_{\alpha\beta}^{\text{SO}}$ to the Kubo formula for the Hall conductivity,

$$\sigma_{\alpha\beta} = \frac{e^2}{\hbar} \sum_n \int_{\text{BZ}} \frac{d^3k}{(2\pi)^3} f_{n\mathbf{k}} \epsilon_{\alpha\beta\gamma} \times \text{Im} \sum_{m \neq n} \frac{\langle n, \mathbf{k} | \hbar v_\beta | m, \mathbf{k} \rangle \langle m, \mathbf{k} | \hbar v_\gamma | n, \mathbf{k} \rangle}{(\varepsilon_{m\mathbf{k}} - \varepsilon_{n\mathbf{k}})^2}. \quad (18)$$

When the density of states at the Fermi level vanishes, the intrinsic anomalous Hall conductivity is derived by the Streda formula [3, 4, 41],

$$\begin{aligned} \sigma_{\alpha\beta} &= -e \epsilon_{\alpha\beta\gamma} \frac{\partial M_\gamma^{\text{orbit}}}{\partial \mu}, \\ &= -e \epsilon_{\alpha\beta\gamma} \frac{\partial \chi_{\gamma\delta}^{\text{SO}}}{\partial \mu} B_\delta^{\text{spin}}. \end{aligned} \quad (19)$$

The topological Dirac semimetals possess time reversal symmetry, so that the Hall conductivity is zero in the absence of the magnetic field. On the other hand, in the presence of the magnetic field, this formula suggests that the anomalous Hall conductivity at the Dirac point becomes finite beside the ordinary Hall conductivity, if $\chi_{\gamma\delta}^{\text{SO}}$ is not symmetric as a function of the Fermi energy ε_F . In the following section, we only consider $\chi_{\alpha\alpha}^{\text{SO}}$, because $\chi_{\alpha\beta}^{\text{SO}}$ ($\alpha \neq \beta$) becomes zero from the view point of the crystalline symmetry in Na_3Bi and Cd_3As_2 .

B. Numerical results

Numerically differentiating Eq. (16) with respect to B_α^{spin} , we obtain $\chi_{\alpha\alpha}^{\text{SO}}$. In Sec. III and IV, we omit $\varepsilon_{\mathbf{k}}$

in Eq. (2) for simplicity. This simplification does not change essential results in the following calculations. In Sec. V, we incorporate $\varepsilon_{\mathbf{k}}$ in order to compare the spin-orbit crossed susceptibility and the spin susceptibility quantitatively in Na_3Bi and Cd_3As_2 . Figure 1 shows the spin-orbit crossed susceptibility χ_{zz}^{SO} at $\varepsilon_F = 0$ as a function of the separation of the Dirac 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 χ_{zz}^{SO} . Figure 1 (a), (b), and (c) show that χ_{zz}^{SO} increases linearly with k_D and satisfy following relation,

$$\chi_{zz}^{\text{SO}} = g_+ \mu_B \frac{2e}{\hbar} \frac{k_D}{\pi}. \quad (20)$$

χ_{zz}^{SO} is proportional to the separation of the Dirac points k_D and the coupling constant $g_+ \mu_B$.

Eq. (20) is given by numerical calculation. This result is understood as follows. χ_{zz}^{SO} is obtained as

$$\chi_{zz}^{\text{SO}} = \int_{-\pi/c}^{\pi/c} \frac{dk_z}{2\pi} \chi_{zz}^{\text{SO}(2D)}(k_z), \quad (21)$$

where $\chi_{zz}^{\text{SO}(2D)}(k_z)$ is the 2D spin-orbit crossed susceptibility at fixed k_z , which is defined in the same way as Eq. (12). $\chi_{zz}^{\text{SO}(2D)}$ is quantized as $2g_+ \mu_B e/\hbar$ in the 2D-QSHI and vanishes in the ordinary insulators [4, 7]. The topological Dirac semimetal is regarded as layers of the 2D-QSHI stacked in the momentum space and the spin Chern number on the k_x - k_y plane with fixed k_z becomes finite only between the Dirac points. As a result, we obtain Eq. (20). The sign of χ_{zz}^{SO} depends on the spin Chern number on the k_x - k_y plane with fixed k_z between the Dirac points. This is analogous to the anomalous Hall conductivity in the Weyl semimetals [2, 23, 42]. In Fig. 1 (d), χ_{zz}^{SO} increases linearly at small k_D but deviates from Eq. (20) for finite γ . This is because the z -component of spin is not conserved in the presence of H_{xy} , Eq. (5), and the above argument for 2D-QSHI is not applicable to the present system. In the following calculation, we set $m_0 = -2m_2$, $m_1 = m_2$, $m_1/t = 1$ and $c/a = 1$.

Figure 2 shows $\chi_{\alpha\alpha}^{\text{SO}}$ at $\varepsilon_F = 0$ as a function of γ . At $\gamma = 0$, χ_{zz}^{SO} is finite as we mentioned above. On the other hand, χ_{xx}^{SO} and χ_{yy}^{SO} are zero. This means that the orbital magnetization is induced only along z -axis, which is the rotational symmetry axis. As a function of γ , χ_{zz}^{SO} is an even function and $\chi_{xx(yy)}^{\text{SO}}$ is an odd function.

Figure 3 (a) shows χ_{zz}^{SO} around the Dirac point as a function of ε_F . When $g_-/g_+ = 0$, χ_{zz}^{SO} is an even function around the Dirac point. At $\varepsilon_F = 0$, χ_{zz}^{SO} is independent of g_-/g_+ as we see it in Fig. 1 (b). When $g_-/g_+ \neq 0$, however, χ_{zz}^{SO} is asymmetric and the derivative of χ_{zz}^{SO} is finite. This suggests that the Hall conductivity is finite when $g_-/g_+ \neq 0$. Calculating Eq. (18) numerically, We confirm that the Hall conductivity is finite at $\varepsilon_F = 0$. Figure 3 (b) shows σ_{xy} as a function g_-/g_+ . σ_{xy} linearly increases with g_-/g_+ . The topological Dirac semimetal

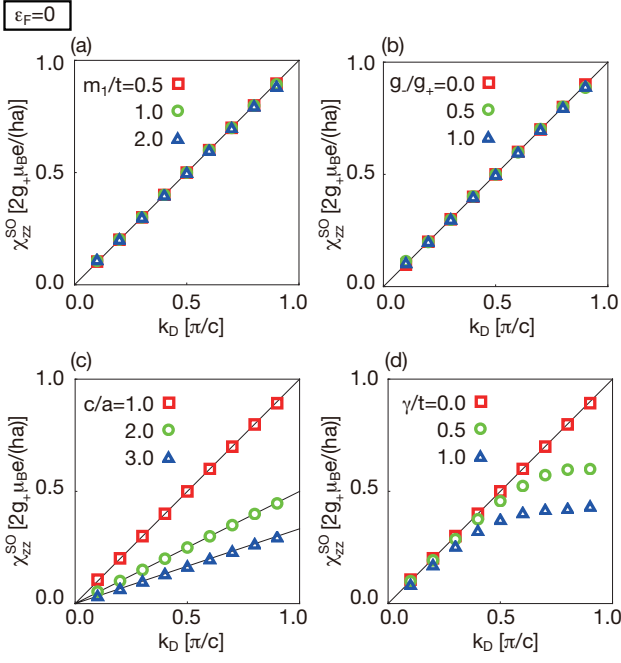


FIG. 1: The spin-orbit crossed susceptibility χ_{zz}^{SO} at $\varepsilon_F = 0$ as a function of k_D . We set the parameters $m_1 = m_2$, $m_1/t = 1$, $g_-/g_+ = 1$, $c/a = 1$, and $\gamma = 0$, if the parameters are not indicated in each figure. The panels (a), (b), and (c) show that χ_{zz}^{SO} is proportional to k_D , which means that χ_{zz}^{SO} reflects the topological property of the electronic structure. From these numerical results, we obtain analytical expression for χ_{zz}^{SO} , Eq. (20), which is independent of model parameters except for k_D and g_+ . The panel (d) show that H_{xy} reduces χ_{zz}^{SO} but it is negligible for sufficiently small k_D .

is viewed as a time reversal pair of the Weyl semimetal with up and down spin. Therefore, the Hall conductivity completely cancel with each other. Even in the presence of g_+ Zeeman term (the symmetric term), the cancellation is retained. In the presence of g_- Zeeman term (the antisymmetric term), on the other hand, the cancellation is broken. This is because g_- Zeeman term changes the separation of the Dirac points and the direction of the change is opposite for the up and down spin Weyl semimetals. As a result, the Hall conductivity is finite in $g_-/g_+ \neq 0$ and given by

$$\sigma_{xy} = \frac{2}{\pi} \frac{e^2}{\hbar a} \frac{g_- \mu_B B^{\text{spin}}}{t}. \quad (22)$$

This expression is quantitatively consistent with the numerical result in Fig. 3 (b).

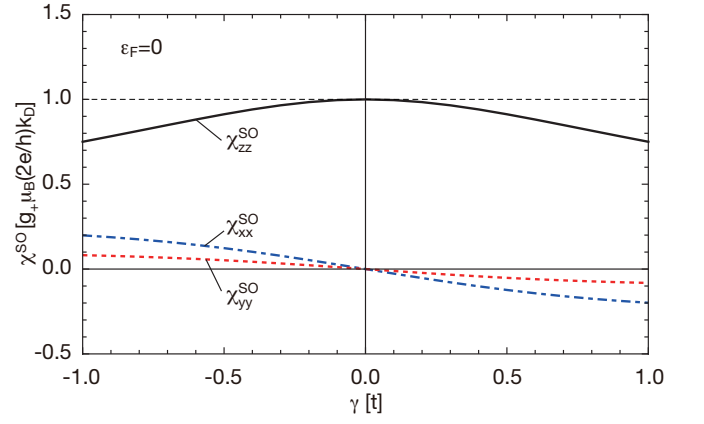


FIG. 2: The spin-orbit crossed susceptibility as a function of γ . The solid black curve is χ_{zz}^{SO} , the blue dashed curve is χ_{xx}^{SO} , and the red dashed curve is χ_{yy}^{SO} . We set the parameters $m_0 = -2m_2$, $m_1 = m_2$, $m_1/t = 1$, $g_-/g_+ = 1$, and $c/a = 1$. Breaking the conservation of σ_z , i.e., with the increase of γ , χ_{zz}^{SO} is reduced, while χ_{xx}^{SO} and χ_{yy}^{SO} become finite.

IV. SPIN SUSCEPTIBILITY

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

$$\chi_{\alpha\alpha}^{\text{spin}}(\mathbf{q}, \varepsilon_F) = \frac{1}{V} \sum_{n\mathbf{k}} \frac{-f_{n\mathbf{k}} + f_{m\mathbf{k}-\mathbf{q}}}{\varepsilon_{n\mathbf{k}} - \varepsilon_{m\mathbf{k}-\mathbf{q}}} \times \mu_B^2 |\langle n, \mathbf{k} | g_+ \tau_0 \sigma_\alpha + g_- \tau_z \sigma_\alpha | m, \mathbf{k} - \mathbf{q} \rangle|^2, \quad (23)$$

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

$$\lim_{|\mathbf{q}| \rightarrow 0} \chi_{\alpha\alpha}^{\text{spin}}(\mathbf{q}, \varepsilon_F) = \chi_{\alpha\alpha}^{\text{intra}}(\varepsilon_F) + \chi_{\alpha\alpha}^{\text{inter}}(\varepsilon_F), \quad (24)$$

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

$$\chi_{\alpha\alpha}^{\text{intra}}(\varepsilon_F) = \frac{1}{V} \sum_{n\mathbf{k}} \left(-\frac{\partial f_{n\mathbf{k}}}{\partial \varepsilon_{n\mathbf{k}}} \right) \times \mu_B^2 |\langle n, \mathbf{k} | g_+ \tau_0 \sigma_\alpha + g_- \tau_z \sigma_\alpha | n, \mathbf{k} \rangle|^2, \quad (25)$$

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

$$\chi_{\alpha\alpha}^{\text{inter}}(\varepsilon_F) = \frac{1}{V} \sum_{n \neq m, \mathbf{k}} \frac{-f_{n\mathbf{k}} + f_{m\mathbf{k}}}{\varepsilon_{n\mathbf{k}} - \varepsilon_{m\mathbf{k}}} \times \mu_B^2 |\langle n, \mathbf{k} | g_+ \tau_0 \sigma_\alpha + g_- \tau_z \sigma_\alpha | m, \mathbf{k} \rangle|^2. \quad (26)$$

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 $\chi_{\alpha\alpha}^{\text{inter}}$ [29]. From the above expression, we see that $\chi_{\alpha\alpha}^{\text{inter}}$ becomes finite, when the matrix elements of the

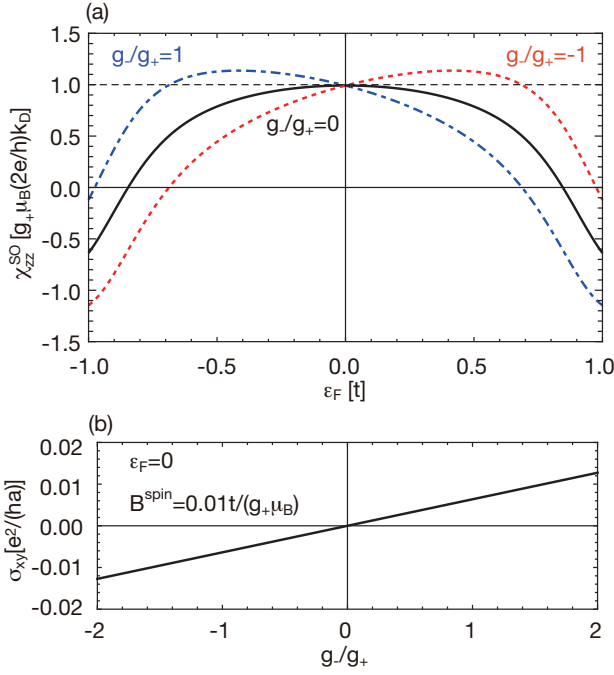


FIG. 3: The spin-orbit crossed susceptibility χ_{zz}^{SO} as a function of ε_F and the Hall conductivity as a function of g_-/g_+ . We set the parameters $m_0 = -2m_2, m_1 = m_2, m_1/t = 1, c/a = 1$, and $\gamma = 0$. At $\varepsilon_F = 0$, the value of χ_{zz}^{SO} is independent of g_- but its ε_F dependence changes at finite g_- . Consequently, the Hall conductivity becomes finite in accordance with Eq. (19).

spin magnetization operator between the conduction and valence bands is non-zero, i.e. the commutation relation between the Hamiltonian and the spin magnetization operator is non-zero. If the Hamiltonian and the spin magnetization operator commute,

$$\langle n, \mathbf{k} | [H_{\mathbf{k}}, g_+ \tau_0 \sigma_x + g_- \tau_z \sigma_z] | m, \mathbf{k} \rangle = 0, \quad (27)$$

the interband matrix element satisfies

$$(\varepsilon_{n\mathbf{k}} - \varepsilon_{m\mathbf{k}}) \langle n, \mathbf{k} | g_+ \tau_0 \sigma_x + g_- \tau_z \sigma_z | m, \mathbf{k} \rangle = 0. \quad (28)$$

This equation means that there is no interband matrix element and $\chi_{\alpha\alpha}^{\text{inter}} = 0$, because $\varepsilon_{n\mathbf{k}} - \varepsilon_{m\mathbf{k}} \neq 0$.

In the following, we set $\varepsilon_F = 0$, where the density of states vanishes. Therefore, there is no intraband contribution and we only consider the interband contribution. We numerically calculate Eq. (26). Figure 4 shows the spin susceptibility $\chi_{\alpha\alpha}^{\text{spin}}$ as a function of (a) γ and (b) g_-/g_+ . In the following, we explain the qualitative behavior of $\chi_{\alpha\alpha}^{\text{spin}}$ using the commutation relation between the Hamiltonian and the spin magnetization operator. In Fig. (4) (a), χ_{zz}^{spin} vanishes at $\gamma = 0$, because the Hamiltonian, H_{TDS} , and the spin magnetization operator of z -component, $g_+ \mu_B \tau_0 \sigma_z$, commute,

$$[H_{\text{TDS}}, g_+ \mu_B \tau_0 \sigma_z] = 0. \quad (29)$$

For finite γ , on the other hand, χ_{zz}^{spin} increases with $|\gamma|$. This is because the commutation relation between H_{xy} and $g_+ \mu_B \tau_0 \sigma_z$ is non-zero,

$$[H_{xy}, g_+ \mu_B \tau_0 \sigma_z] \neq 0, \quad (30)$$

and χ_{zz}^{inter} gives finite contribution. χ_{xx}^{spin} and χ_{yy}^{spin} are finite even in the absence of H_{xy} , i.e. $\gamma = 0$, because H_{TDS} and $g_+ \mu_B \tau_0 \sigma_\alpha$ ($\alpha = x, y$) do not commute,

$$\begin{aligned} [H_{\text{TDS}}, g_+ \mu_B \tau_0 \sigma_x] &\neq 0, \\ [H_{\text{TDS}}, g_+ \mu_B \tau_0 \sigma_y] &\neq 0. \end{aligned} \quad (31)$$

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

$$[H_{\text{TDS}}, g_- \mu_B \tau_z \sigma_z] \neq 0. \quad (32)$$

Consequently, χ_{zz}^{inter} gives finite contribution, though the z -component of spin is a good quantum number. The antisymmetric term does not break the four-fold rotational symmetry along z -axis, so that χ_{xx}^{spin} is equal to χ_{yy}^{spin} in Fig. (4) (b).

The spin susceptibility $\chi_{\alpha\alpha}^{\text{spin}}$ is also anisotropic but contrasts with the spin-orbit crossed susceptibility $\chi_{\alpha\alpha}^{SO}$. χ_{xx}^{spin} and χ_{yy}^{spin} are larger than χ_{zz}^{spin} , in contrast χ_{zz}^{SO} is larger than χ_{xx}^{SO} and χ_{yy}^{SO} . Therefore, the angle dependence measurement of magnetization will be useful to separate the contribution from the each susceptibility.

V. DISCUSSION

In this section, we quantitatively compare the spin-orbit crossed susceptibility χ_{zz}^{SO} and the spin susceptibility χ_{zz}^{spin} at the Dirac points as a function of g_-/g_+ . In the following calculation, we set the parameters to reproduce the energy band structure around the Γ point calculated by the first principle calculation for Cd_2As_3 and Na_3Bi [10, 15]. The parameters are listed in the table and we omit H_{xy} , i.e. $\gamma = 0$.

Figure 5 shows the two susceptibilities as a function of g_-/g_+ . We find that the two susceptibilities are approximately written as

$$\chi_{zz}^{\text{spin}} \sim \left(\frac{g_-}{g_+} \right)^2, \quad (33)$$

and

$$\chi_{zz}^{SO} \sim -\frac{1}{g_+} \left(\chi_0 + \frac{g_-}{g_+} \right), \quad (34)$$

by numerical fitting. In the present parameters, χ_{zz}^{SO} is negative and depends on g_-/g_+ . The dependence on

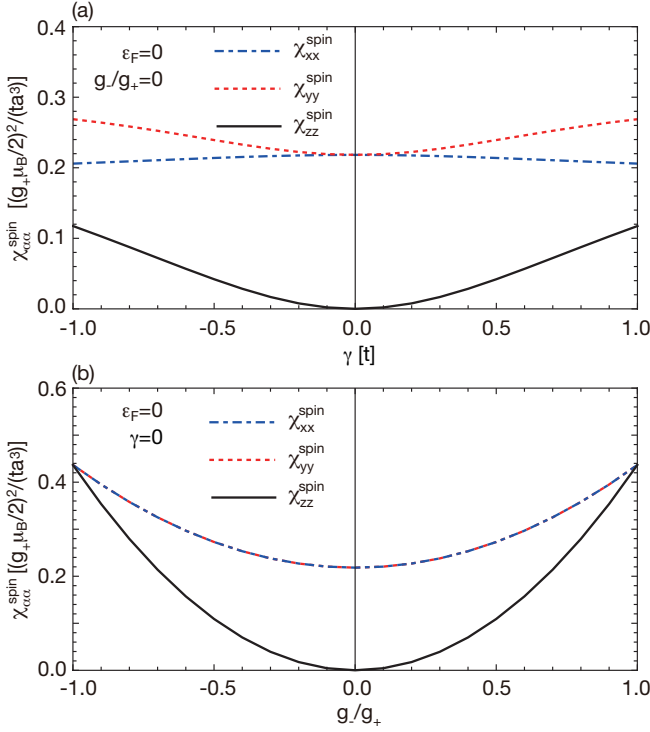


FIG. 4: The spin susceptibility $\chi_{\alpha\alpha}^{\text{spin}}$ at $\varepsilon_F = 0$ as a function of (a) γ and (b) g_-/g_+ . We set $m_0 = -2m_2$, $m_1 = m_2$, $m_1/t = 1$, and $c/a = 1$. At $\gamma = 0$ and $g_-/g_+ = 0$, $\chi_{zz}^{\text{spin}} = 0$ while $\chi_{xx}^{\text{spin}}, \chi_{yy}^{\text{spin}} > 0$. These behaviors are explained by the commutation relation between the Hamiltonian and the spin magnetization operators as discussed in the main text.

g_-/g_+ originates from the existence of $\varepsilon_{\mathbf{k}}$, which breaks the particle-hole symmetry. The g -factors are experimentally estimated as $g_s = 18.6$ for Cd_2As_3 [43] and $g_- = 20$ for Na_3Bi [44]. Unfortunately, there is no experimental data which determines both of g_s, g_p or g_+, g_- . From Fig. 5, we see that χ_{zz}^{SO} can dominate over χ_{zz}^{spin} if $g_-/g_+ \simeq 0$. As far as we know, there is no experimental observation of the magnetic susceptibility in these materials. We expect the experimental observation in near future and our estimation of χ_{zz}^{SO} will be useful to appropriately analyze experimental data.

Material parameters		
	Cd_3As_2	Na_3Bi
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[Å]	5.07[Å]
c	25.43[Å]	9.66[Å]

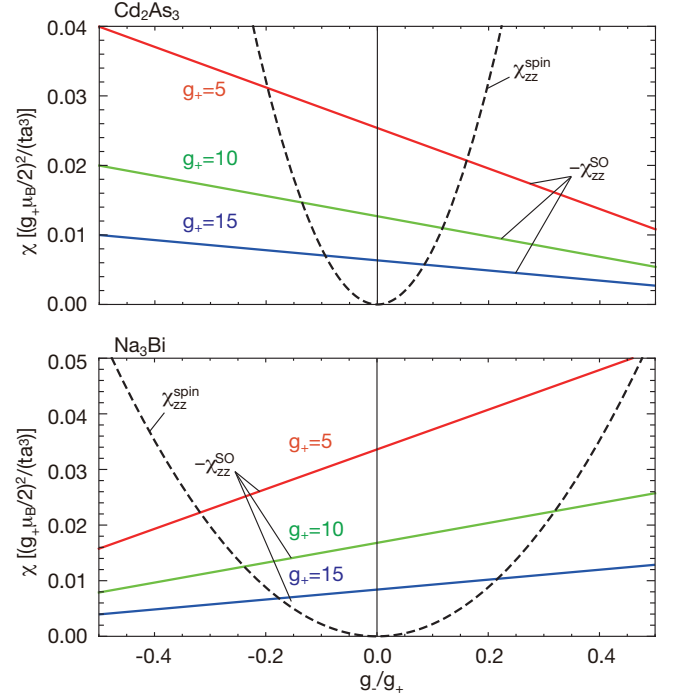


FIG. 5: The spin-orbit crossed susceptibility χ_{zz}^{SO} and the spin susceptibility χ_{zz}^{spin} at the Dirac points as a function of g_-/g_+ . The dashed curve is χ_{zz}^{spin} and the solid lines are χ_{zz}^{SO} . The upper (lower) panel shows Cd_2As_3 (Na_3Bi). When g_-/g_+ are sufficiently small, χ_{zz}^{SO} becomes comparable to χ_{zz}^{spin} .

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. This means that χ_{zz}^{SO} reflects topological property of the electronic structure. The spin-orbit crossed susceptibility is induced only along the rotational symmetry axis. We also calculate 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_2As_3 and Na_3Bi . At the Dirac point, the orbital susceptibility logarithmically diverges and gives dominant contribution to the total susceptibility. Off the Dirac point, on the other hand, the orbital susceptibility decreases [6, 24, 25] and the contribution from the spin susceptibility and the spin-orbit crossed susceptibility is important for appropriate estimation of the total susceptibility.

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