A spin current has novel linear and second-order nonlinear optical effects due to its symmetry properties. With the symmetry analysis and the eight-band microscopic calculation we have systematically investigated the interaction between a spin current and a polarized light beam (or the "photon spin current") in direct-gap semiconductors. This interaction is rooted in the intrinsic spin-orbit coupling in valence bands and does not rely on the Rashba or Dresselhaus effect. The light-spin current interaction results in an optical birefringence effect of the spin current. The symmetry analysis indicates that in a semiconductor with inversion symmetry, the linear birefringence effect vanishes and only the circular birefringence effect exists. The circular birefringence effect is similar to the Faraday rotation in magneto-optics but involves no net magnetization nor breaking the time-reversal symmetry. Moreover, a spin current can induce the second-order nonlinear optical processes due to the inversion-symmetry breaking. These findings form a basis of measuring a pure spin current where and when it flows with the standard optical spectroscopy, which may provide a toolbox to explore a wealth of physics connecting the spintronics and photonics.

Detection of spin currents is important for characterizing and applications in future spintronics technologies. While a polarized spin current may be detected by the conventional Faraday/Kerr rotation spectroscopy or through ferromagnetic filters a pure spin current, without a direct electromagnetic induction, is much less traceable. Still, pure spin currents have been detected in a few pioneering experiments in which they were converted into signals detectable by conventional techniques. For example, the spin-polarized electrons or excitons accumulated at the sample edges where a spin current is terminated may be detected by the Faraday/Kerr rotation or polarized light emission. Or the inverse spin Hall effect can be used to covert a spin current into charge/voltage signals for electric measurement. All such measurements, however, disturb the spin currents to some extent and are indirect. We are motivated to find a non-destructive way to directly measure a pure spin current by mapping the photon polarization into a spin-1/2 in the Jones vector representation

$$\cos \frac{\theta}{2} e^{i\phi/2} n_+ + \sin \frac{\theta}{2} e^{-i\phi/2} n_- \sim |\theta, \phi\rangle,$$  

where the right/left circular polarization $n_+/-$ corresponds to the spin up/down state $|\uparrow, \downarrow\rangle$ quantized along the light propagation direction. The effective interaction between a pure spin current and a polarized light causes a phase delay which depends on the light polarization and wavevector. The observable result is a circular birefringence effect which is similar to the Faraday rotation but involves no net magnetization nor time-reversal symmetry breaking. Since Faraday’s discovery in 1845 the circular birefringence effect of spin currents is the first example of Faraday rotation without time-reversal symmetry breaking. We dub this effect as spin current Faraday effect.

At this point, we should mention a recent remarkable experiment realizing the direct in-situ detection of a spin current through the Doppler effect of a spin-wave. In fact, the observed Doppler effect and our predicted optical birefringence effect are fundamentally related to each other. The former is the frequency shift of the spin wave, while the latter is the phase shift of the light accumulated by a frequency shift over a coupling time. The frequency shift is measured in the near-field as in the experiment, while the phase shift should be measured in the far field by light polarization detection. Fundamentally, both are due to the effective coupling between a pure spin current and another “probe spin current”, either a spin wave or a polarized light, mediated by virtual excitations in the systems.

The effective light-spin current interaction is induced in a semiconductor by virtual excitations of electron-hole pairs. The specific form of the phenomenological coupling depends
on the microscopic mechanisms. Since the light polarization essentially couples only to the orbital motion of electrons, the spin-orbit interaction is needed to establish the effective coupling. As there is inherent spin-orbit coupling in the valence bands due to the relativity effect, the Rashba or Dresselhaus effect due to the spatial inversion asymmetry is not a necessity, thus the system can bear the inversion symmetry. The optical birefringence effect of spin currents is usually very weak, because a tiny light wave vector \( \mathbf{q} \) is involved in the coupling to the velocity \( \mathbf{v} \) of the pure spin current. However, if the velocity of spin currents couples to another optical field, \( \mathbf{q} \cdot \mathbf{v} \Rightarrow \mathbf{F}_{\mathbf{z}} \cdot \mathbf{v} \)

the coupling will be much enhanced. This means we can use the second optical field to drive the spins, which may result in the nonlinear optics of the pure spin current. In fact, such an analogy stimulated the prediction of the second-order nonlinear optical effects of pure spin currents which was soon verified by experiments.

In Refs. [20] and [21] we have sketched the main ideas based on symmetry arguments and given the key expressions in a special model neglecting the energy band anisotropy. In this paper, we will investigate in a more comprehensive way the linear and second-order nonlinear optical effects of pure spin currents, including a systematic symmetry analysis of all relevant physical quantities, and a detailed derivation for the effective Hamiltonian as well as the second-order nonlinear optical susceptibility. The microscopic derivation confirms the qualitative results obtained by the symmetry analysis. In particular, both the symmetry analysis and the microscopic calculation lead to the conclusion that the linear birefringence effect (similar to the Voigt effect in magneto-optics) always vanishes and only the circular birefringence effect exists, and the energy band anisotropy induces only a relatively small quantitative modification of the results. The absence of the Voigt effect is fundamentally related to the lack of the \( |0\rangle \) state in the physical spin of photons [not the pseudo-spin in Eq. (1)]. The microscopic mechanism of both linear and second-order nonlinear effects can be understood in a unified physical picture.

In this paper, we assume that the host semiconductor system has the inversion symmetry. We note that in compound semiconductors such as GaAs the inversion symmetry is broken, which, though a small effect, is critical to some schemes of spin current generation. In our present scheme, however, the small inversion asymmetry in compound semiconductors is not important. For conditions used in our microscopic calculation, the spin splitting resulting from the Dresselhaus effect due to the bulk inversion asymmetry (The Dresselhaus splitting is \( \sim 0.01 \) meV in GaAs with doping density \( \sim 10^{16} \) cm\(^{-3}\)), much less than the detuning of the light from the interband transitions that mediate the effective interaction, so we can neglect the bulk inversion asymmetry in the measurement process even though it could be of vital importance in generating the spin current. Also, in this paper, we consider only bulk materials, so the structure inversion asymmetry plays no role, though it is the basis of the Rashba effect. Without considering the Dresselhaus and Rashba effects due to inversion asymmetries, we avoid the subtlety in the definition of a spin current. The effect of inversion asymmetries on the interaction between the polarized light beams and a spin current, of course, is worth further study, but we prefer leaving this question open in this paper.

The paper is organized as follows. Sec. II presents a systematic symmetry analysis for the coupling system to give a qualitative understanding of the linear and circular birefringence effects and the second-order nonlinear optical effect of pure spin currents. Sec. III gives the theoretical model and microscopic derivations for both the linear and the second-order nonlinear optical effects, and also explains the physical pictures for the microscopic mechanism of optical effects of spin currents. Sec. IV presents the numerical results and discussions of the experiment scheme. Sec. V concludes this paper.

II. SYMMETRY ANALYSIS

We will particularly consider the time-reversal (\( T \)) and the space-inversion (\( P \)) symmetries of all the relevant physical quantities, and the geometry symmetry for a specific form of spin currents. According to the symmetry analysis, a pure spin current may result in a circular birefringence effect but not a linear birefringence effect, and as it breaks \( P \) symmetry, a spin current can induce the second-order nonlinear optical processes.

A. Linear optical effects

We assume the whole system has the \( T \) and \( P \) symmetries at equilibrium. Namely, the effective coupling between a spin polarization or a spin current in the semiconductor system and a probe should have both symmetries, i.e., the transformation properties of the effective Hamiltonian \( \mathcal{H}_{\text{eff}} \) are

\[
\begin{pmatrix}
T & P \\
\mathcal{H}_{\text{eff}} & + & +
\end{pmatrix}
\]

where +/- refers to even/odd under the corresponding symmetry transformations.

In our study, a pure spin current is made of a non-equilibrium distribution of spin polarization in the momentum space. In general, it can be quantified by a rank-2 pseudo-tensor defined by (with volume of the material taken as unity)

\[ J = \sum_{p} j_{p} = e \sum_{p} s_{p} v_{p}, \]  

where \( s_{p} \) is the spin polarization and \( v_{p} \) is the velocity of a particle with wave vector \( p \), and \( e \) is the electron charge. The "photon spin current" tensor for a polarized light beam with electric field \( \mathbf{F}(r, t) = (F_{+} \mathbf{n}_{+} + F_{-} \mathbf{n}_{-}) e^{i q \cdot \mathbf{r} - \omega t} + \text{c.c.} \) is formulated as

\[
\mathbb{I} = \mathbf{I}_{\mathbf{q}} \equiv q (I_{x} \mathbf{xz} + I_{y} \mathbf{yz} + I_{z} \mathbf{zz}),
\]

\[
I_{j} = \frac{1}{2} \sum_{\mu, \nu = x, y, z} \epsilon_{\mu \nu z} F_{\mu} F_{\nu},
\]
where \( \mathbf{q} \) is the wave vector of the light beam, the unit vector \( \mathbf{z} \) is chosen along the direction of \( \mathbf{q} \) so that \( \mathbf{q} = q \mathbf{z} \), the unit vectors \( \mathbf{x} \) and \( \mathbf{y} \) are related to the light polarization through \( \mathbf{n}_s \equiv (\pm \mathbf{x} - i \mathbf{y}) / \sqrt{2} \), and \( \sigma^j (j = x, y, z) \) is the Pauli matrix. For completeness, we also consider the spin polarization of the system

\[
S = \sum_p s_p. 
\]  

(5)

The transformation properties under the \( \mathcal{T} \) and \( \mathcal{P} \) of the relevant physical quantities are

\[
\begin{array}{cccc}
\mathbf{S} & \mathbf{q} & l_x, l_z & l_y \\
\mathcal{T} & - & - & + \\
\mathcal{P} & + & - & + \\
\end{array}
\]

(6)

It is worth mentioning here that there is no \( |0\rangle \) state in the physical spin of photons, and the photon pseudo spin \( l_x \) and \( l_z \) do not break the \( \mathcal{T} \)-symmetry, for it involves the 2nd-order spin flip processes such as \( |+1\rangle \rightarrow |0\rangle \rightarrow |-1\rangle \). In the following we will use these quantities to form an effective Hamiltonian (undetermined up to a few coupling constants) satisfying the \( \mathcal{T} \) and \( \mathcal{P} \) symmetries. Since the interaction of the light with a spin is usually weak, we only consider the effect in the leading order, which is bilinear in the spin and light quantities.

**Net spin polarization.** The only optical quantity of the same symmetry-breaking type as the spin polarization is \( l_z \mathbf{z} \). Thus the effective interaction between a spin polarization and a light beam has the form

\[
\mathcal{H}^{(0)}_{\text{eff}} = \zeta_0 l_z \mathbf{S} \cdot \mathbf{z},
\]

(7)

with a coupling constant \( \zeta_0 \) to be determined by the specific microscopic mechanism. Such a coupling corresponds to the conventional Faraday effect in magnetooptics. We would like to point out here that a spin polarization could induce the Voigt effect. In order to have the same symmetry-breaking type for \( l_x \) and \( l_z \), the spin polarization should be of an even power. Thus to the leading order, the effective interaction has the form

\[
\mathcal{H}_{\text{eff}}^{\text{Voigt}} = \zeta_0^{\text{Voigt}} l_x (\mathbf{S} \cdot \mathbf{x})^2.
\]

(8)

This explains that the Voigt effect is quadratic in the spin polarization or the applied external magnetic field.

**Pure spin current.** There is no term in the light polarization \( l_j (j = x, y, z) \) that has the same symmetry-breaking type as the spin current, so it is not possible to have linear (nonlinear optics is of course possible) interaction between the spin current and the light without involving the wave vector. Considering the wave vector of the light between the spin current and the photon current \( ql \mathbf{z} \mathbf{z} \) is possible. The linear birefringence effect (similar to the Voigt effect in magnetooptics) is absent. Due to the lack of \( |0\rangle \) state in the physical spin of photons, \( l_x \) and \( l_z \) preserve \( \mathcal{T} \) symmetry. Therefore there is no linear coupling of \( l \) to \( ql \mathbf{xx} \) and \( ql \mathbf{yy} \).

Furthermore, if the system has spherical symmetry, the effective Hamiltonian would have a simple tensor contraction form as

\[
\mathcal{H}_{\text{eff}}^{(1)} = \zeta_1 ql \mathbf{Tr} (\mathbf{J}) + \zeta_2 ql \mathbf{z} \cdot \mathbf{J} \cdot \mathbf{z},
\]

(9)

with only two coupling constants \( \zeta_1 \) and \( \zeta_2 \) to be determined by the microscopic mechanisms. A possible spherical symmetric system is the vacuum, but in general a semiconductor as a crystal does not have this symmetry. The general effective interaction in a semiconductor should have the form

\[
\mathcal{H}_{\text{eff}}^{(1)} = ql \mathbf{zz} : \mathcal{A} : \mathbf{J},
\]

(10)

where \( \mathcal{A} \) is a parameter tensor determined by the microscopic structure of the material. Since only the light polarization term \( l_z \) appears in the interaction, the optical birefringence effect is circular, similar to the Faraday rotation.

In realistic case, the spin current often has some special form. As a general case a spin current tensor can have the form as

\[
\mathbf{j} = J_x \mathbf{XZ} + J_y \mathbf{YZ} + J_z \mathbf{ZZ} = \mathbf{JZ},
\]

(11)

where \( \mathbf{Z} \) is the unit vector along the direction of spin current, the unit vectors \( \mathbf{X} \) and \( \mathbf{Y} \) are perpendicular to \( \mathbf{Z} \), and \( \mathbf{J} \) denotes the spin current amplitude vector, which is an axial vector parallel to the spin polarization direction. Now the \( \mathbf{z} \) and \( \mathbf{Z} \) axes form a special plane. If the system has reflection symmetry with respect to this plane (e.g., the system is spherically symmetric or the plane is along a special crystal direction of the semiconductor), the symmetry properties of the relevant quantities under reflection with respect to the \( \mathbf{z} \cdot \mathbf{Z} \) plane will impose further constraint on the interaction and significantly simplify the Hamiltonian. Under the reflection, the relevant quantities transform as

\[
\begin{array}{ccc|ccc}
\text{Reflection with } \mathbf{z} \cdot \mathbf{Z} \text{ plane} & q & l_x & l_y & l_z & + & - \\hline
\end{array}
\]

(12)

where \( \mathbf{J}_i \) is the component of \( \mathbf{J} \) in the plane and \( \mathbf{J}_\perp \) is the perpendicular component. By the table above, it is evident that to keep the effective Hamiltonian invariant only the in-plane component of \( \mathbf{J}_i \) would couple with the \( ql \). Without loss of generality, let \( \mathbf{Y} \) be perpendicular to the \( \mathbf{z} \cdot \mathbf{Z} \) plane, the effective Hamiltonian reads

\[
\mathcal{H}_{\text{eff}}^{(1)} = A_1 ql l_x Z + A_2 ql l_z X,
\]

(13)

in which two coupling constants \( A_1 \) and \( A_2 \) are to be determined by microscopic calculation. Alternatively, the Hamiltonian can be expressed in a form independent of the choice of the \( \mathbf{X} \) and \( \mathbf{Y} \) axes as

\[
\mathcal{H}_{\text{eff}}^{(1)} = \zeta_1 ql l_z Z + \zeta_2 ql l_x \mathbf{z} \cdot \mathbf{J} \cdot \mathbf{z},
\]

(14)

which is the same as Eq. (9), but does not require the spherical symmetry of materials.

The physical effect of the effective coupling can be extracted from the linear optical susceptibility,

\[
\chi_{\mu, \nu} + \chi_{\nu, \mu}^* = (1/\epsilon_0) \partial^\mu \mathcal{H}_{\text{eff}} / (\partial F^\mu_\rho \partial F^\rho_\nu),
\]

(15)

where \( \epsilon_0 \) is the vacuum permittivity. Thus we get an opposite susceptibility for opposite circular polarization in presence of a spin polarization or a pure spin current

\[
\begin{align}
\chi_{+ +}^{(0)} & = - \chi_{--}^{(0)} = (1/4\epsilon_0) \zeta_0 \mathbf{z} \cdot \mathbf{S}, \\
\chi_{+ +}^{(1)} & = - \chi_{--}^{(1)} = (q/4\epsilon_0) (\zeta_1 J_Z + \zeta_2 J_z \cdot \mathbf{J} \cdot \mathbf{z}).
\end{align}
\]
The effective energy shift resulting from the light-spin or light-spin current interaction means a phase shift in the light observed in the far-field. Eq. (16a) is nothing but the conventional Faraday rotation in magneto-optics, 

\[
\chi^{(2)}_{\text{Faraday}} = J_\text{XY} (\chi_1 \text{XXX} + \chi_2 \text{XYX} + \chi_3 \text{YXY} + \chi_4 \text{ZZY} + \chi_5 \text{XYZ} + \chi_6 \text{YYZ} + \chi_7 \text{YYY}) ,
\]

with seven independent parameters to be determined. Similar symmetry analysis can be applied to \( J_\text{YZ} \). Such unique polarization dependence of the second-order optical susceptibility can be used to distinguish the longitudinal and transverse components of a spin current, and also to single out the spin-current signature from the effects of the material background or a charge current. 

### III. MICROSCOPIC CALCULATION

To quantitatively determine the linear and the second-order nonlinear optical effects of a spin current, we will perform the microscopic calculation for a pure spin current in a bulk direct-gap semiconductor using the standard perturbation theory. 

We as-
The spin current, which results from the non-equilibrium distribution of CB electrons, is expressed by Eq. (3), where the velocity and the spin polarization of an electron with momentum \( \mathbf{p} \) is respectively given by \( \mathbf{v}_p = \nabla_p E_{p} \) and \( \mathbf{s}_p = (1/2) \sum_{\mu\nu} \sigma_{\mu\nu} f_{\mu\nu, p} \) with \( \sigma \) denoting the Pauli matrices.

### B. Linear optical effects

The direct interaction between a light beam and a semiconductor is the dipole interband optical transitions. Only through the spin-orbit coupling in valence bands, may the light beam interact with the spin of electrons.

For the dipole interband transition \([\text{Fig. 1(b)}] \& \text{(c)}\), the polarization density operator reads \([\text{31}]\)

\[
\hat{\mathbf{P}}_{\text{inter}} (\mathbf{r}) = -d_{cv} \sum_{k,p,\mu\pm} \left( \mathbf{n}_{\mathbf{p} k} \hat{\mathbf{p}}_{\mu \pm} \mathbf{e}_{\mu \pm, k} + \frac{1}{\sqrt{3}} \mathbf{n}_{\mathbf{p} k} \hat{\mathbf{p}}_{\mu \pm} \mathbf{e}_{\mu \pm, k} \right)

- \sqrt{\frac{2}{3}} z_{p} \hat{\mathbf{p}}_{\mu \pm} \mathbf{e}_{\mu \pm, k} - \mu \sqrt{\frac{2}{3}} z_{p} \hat{\mathbf{p}}_{\mu \pm} \mathbf{e}_{\mu \pm, k}

+ \sqrt{\frac{\mu}{\sqrt{3}}} z_{p} \hat{\mathbf{p}}_{\mu \pm} \mathbf{e}_{\mu \pm, k} e^{i \mathbf{p} \cdot \mathbf{r} - i \mathbf{k} \cdot \mathbf{r}} + \text{H.c.},
\]

where \( \mathbf{n}_{\mathbf{p} k} \equiv \mp (\mathbf{x}_p \pm i \mathbf{y}_p) / \sqrt{2} \) denotes the right/left circular polarization about the momentum direction \( \mathbf{p} \), \( z_{p} \equiv \mathbf{p} / \mu \), and \( \hat{\mathbf{d}}_{cv} \equiv -d_{cv} \). As will be discussed in Sec. [IVB], the momentum dependence of the dipole moment has no significant effect, so here we assume the interband dipole moment \( d_{cv} \) independent of the momentum. With the dipole interaction with a light \( H_1 (t) = -\int \mathbf{F}_{\text{inter}} (\mathbf{r}) \cdot \mathbf{F} (\mathbf{r}, t) d\mathbf{r} \), the light-matter interaction Hamiltonian in the rotating wave approximation can be explicitly expressed as

\[
\hat{H}_1 = \exp \left( \sum_{\mu,k} \frac{i \omega_{q} \hat{e}_{\mu, k}}{\mu} \right) \hat{H}_0 (t) \exp \left( \sum_{\mu,k} -\frac{i \omega_{q} \hat{e}_{\mu, k}}{\mu} \right)

= d_{cv} \sum_{\mu, \nu, p} \int \mathbf{n}_\nu^* \mathbf{n}_{\mu, p} \hat{\mathbf{p}}_{\mu, q, p} \hat{\mathbf{p}}_{\nu, q, p} + \frac{1}{\sqrt{3}} \mathbf{n}_{\mu, p} \hat{\mathbf{p}}_{\mu, p} \mathbf{e}_{\mu, q, p}

- \sqrt{\frac{2}{3}} z_{p} \hat{\mathbf{p}}_{\mu, \pm} \mathbf{e}_{\mu, q, p} - \mu \sqrt{\frac{2}{3}} z_{p} \hat{\mathbf{p}}_{\mu, \pm} \mathbf{e}_{\mu, q, p}

+ \frac{\mu}{\sqrt{3}} z_{p} \hat{\mathbf{p}}_{\mu, \pm} \mathbf{e}_{\mu, q, p} + \text{H.c.}.
\]

Under the condition that the optical interaction strength is much smaller than the detuning of the light from the valence
band to the Fermi level (the perturbation regime), the effective energy due to the dipole interaction can be derived by the second-order perturbation as

$$\mathcal{H}_{\text{eff}} = \text{Tr} \left[ \hat{\rho}_0 \hat{H}_1 \left( \hat{H}_0 - \omega_q \right)^{-1} \hat{H}_1 \right].$$  \hspace{2cm} (26)

Such effective coupling between a spin current and a polarized light beam on the one hand can be regarded as the frequency shift of the light in the presence of the spin current, and on the other hand can be considered as the energy change of the semiconductor system under the driving of light beam. The second-order perturbation means that there are two virtual optical transitions induced by the electric field of the light: one creating an electron-hole pair and one annihilating the electron-hole pair. The virtual excitations cause no real optical absorption but a phase shift, indicating that the effective coupling is real. The optical effect of the spin-current can be understood as the Pauli blocking in the transition involving different spin states. With this picture in mind, the following microscopic calculation, though lengthy, is quite transparent.

1. Physical Picture

The physical picture for the microscopic mechanism of the spin current Faraday effect is rooted in the fact that a spin will induce a Faraday rotation like a magnet. In Faraday rotation, a linearly polarized optical field $\mathbf{F}$ induces a polarization as a rotation about the spin,

$$\mathbf{P}^{(1)} \propto \frac{\mathbf{F} \times \mathbf{s}_k}{\omega - E_k},$$  \hspace{2cm} (27)

where $\mathbf{s}_k$ is the spin polarization associated with the state of $\mathbf{k}$, $E_k$ is the resonant optical transition energy. This naturally explains Faraday rotation due to spin polarization as in Eq. (26a).

For a pure spin current, we first consider only a pair of spins, $\mathbf{s}_k$ at momentum $\mathbf{k}$, and $-\mathbf{s}_k$ at momentum $-\mathbf{k}$ in the CB [Fig. 2]. This pair can be viewed as a generator of pure spin current. $\mathbf{s}_k$ gives rise to a Faraday rotation of $\mathbf{P}^{(1)}_k \propto \mathbf{F} \times \mathbf{s}_k / (\omega - E_k)$; while $-\mathbf{s}_k$ leads to a Faraday rotation of $\mathbf{P}^{(1)}_{-k} \propto \mathbf{F} \times \mathbf{s}_{-k} / (\omega - E_{-k})$. Therefore, the Faraday rotations caused by the pair of spins cancel each other in the vertical optical transition. However, when the effect of the small light-momentum is taken into consideration, the excitation energy at $\pm \mathbf{k}$ will shift respectively to $E_{\pm k} \sim E_{\pm k} \pm \mathbf{q} \cdot \mathbf{v}_k$ [Fig. 2], and $\pm \mathbf{s}_k$ will induce different Faraday rotations due to opposite energy variation. Up to first order of $\mathbf{q}$, the polarization is $\mathbf{P}^{(1)} \propto \mathbf{F} \times \mathbf{s}_k \mathbf{v}_k / (\omega - E_{\pm k})^2$, where $\mathbf{e}_k \mathbf{v}_k$ is just the spin current tensor contributed by the pair of electrons. This explains the $\mathbf{q}$-dependence of the spin current Faraday effect.

More generally, the hole state wavefunction is also changed when considering the light momentum, which causes extra Berry phase effects [terms proportional to $1/E_F$ in Eq. (27)].

2. Effective Hamiltonian by SO-CB transitions

To better understand the microscopic mechanism of the light-spin current coupling, let us first derive the effective Hamiltonian contributed solely by the transitions between the CB and SO bands. The SO band electrons has 2-fold degeneracy, and the spin states as well as the selection rules for the interband transitions, like the CB electrons, are independent of the momentum [Fig. 1c].

We first consider a single electron with momentum $\mathbf{k}$ and spin polarization $\mathbf{s}_k$. The spin current contributed by this electron is $\mathbf{J}_k = e \mathbf{s}_k \mathbf{v}_k$ with the velocity $\mathbf{v}_k = \mathbf{k}/m_e$. It is convenient to define the spin basis states along the spin polarization direction. In such chosen basis, the spin density matrix of the electron is diagonal. With the population in the spin-up and spin-down states denoted as $f_+$ and $f_-$, respectively, the spin polarization is $\mathbf{s}_k = (f_+ - f_-)/2$. The interband transitions $|1/2, \pm 1/2\rangle_k \leftrightarrow |\pm \rangle_k$ couple to a field with circular polarization $\mp (\mathbf{e}_1 \pm i \mathbf{e}_2)/\sqrt{3}$, and the vertical inter-band transitions $|1/2, \pm 1/2\rangle_k \leftrightarrow |\pm \rangle_k$ couple to a field of linear polarization $\mathbf{e}_3/\sqrt{3}$, where the coordinate system is so defined that $\mathbf{e}_3$ is along the spin polarization direction of the electron considered. Summing up all possible inter-band transitions, the energy shift of this electron due to coupling to an optical field $\mathbf{F}$ is

$$\mathcal{H}_{\text{eff}}^{SO} = -\frac{1}{3} |\mathbf{d}\rangle |\mathbf{d}\rangle^\dagger \sum_\pm \frac{(1 - f_k) \mathbf{F} \cdot (\mathbf{e}_1 \mp i \mathbf{e}_2) (\mathbf{e}_1 \mp i \mathbf{e}_2)^* \mathbf{F}}{\omega_q - E_{\pm k} + \mathbf{q} \cdot \mathbf{F}} - \frac{1}{3} |\mathbf{d}\rangle |\mathbf{d}\rangle^\dagger \sum_\pm \frac{(1 - f_k) \mathbf{F} \cdot \mathbf{e}_3 \mathbf{e}_3^* \mathbf{F}}{\omega_q - E_{\pm k} + \mathbf{q} \cdot \mathbf{F}},$$  \hspace{2cm} (28)

where the factor $(1 - f_k)$ accounts for the Pauli blocking of the interband transitions. The second term, which is related to vertical transitions caused by a linearly polarized field, does
not depend on the spin polarization, so it can be dropped as the background. With expansion to the first order of $\mathbf{q}$ and omission of the background terms, the energy shift becomes

$$
\mathcal{H}_{\text{eff},k}^{SO} = \frac{2}{3} |\mathbf{d}_c|^2 \frac{\mathbf{F} \cdot (\mathbf{e}_1 \mathbf{e}_2 - \mathbf{e}_2 \mathbf{e}_1) \cdot \mathbf{F}}{\omega_q - E_{l,-k} - E_{ek}} + \frac{2}{3} |\mathbf{d}_c|^2 \frac{\mathbf{F} \cdot (\mathbf{e}_1 \mathbf{e}_2 - \mathbf{e}_2 \mathbf{e}_1) \cdot \mathbf{k}}{m(\omega_q - E_{l,-k} - E_{ek})^2}.
$$

(29)

Since $(\mathbf{e}_1 \mathbf{e}_2 - \mathbf{e}_2 \mathbf{e}_1) \cdot \mathbf{F} = \mathbf{F} \times (\mathbf{e}_1 \times \mathbf{e}_2) = \mathbf{F} \times \mathbf{e}_3$, the physical meaning of this coupling is transparent: the linear-polarized optical field will tilt about the spin, which is essentially the Faraday rotation with spin playing the role of a magnet. The summation over the momentum space gives

$$
\mathcal{H}_{\text{eff}}^{SO} = -\frac{4}{3} |\mathbf{d}_c|^2 \frac{1}{\Delta_l} \mathbf{I} \cdot \mathbf{z} - \frac{4}{3} |\mathbf{d}_c|^2 \frac{m_e}{e m \Delta_l} q \mathbf{I} \cdot \mathbf{z} \cdot \mathbf{z}.
$$

(30)

where $\Delta_l$ is the light detuning from SO band to the Fermi surface.

3. Effective coupling by transitions between HH/LH and CB

If the HH bands and LH bands are assumed degenerate, the quantization direction of the 3/2-spin of the HH and LH can be chosen arbitrarily and the effective Hamiltonian are obtained in a similar way to that contributed by the SO-CB transition. However, with the HH-LH splitting considered, the quantization direction of the hole states depends on its moment, thus, with the trivial background omitted, the effective Hamiltonian can be derived explicitly as

$$
\mathcal{H}_{\text{eff}}^{HL} = |\mathbf{d}_c|^2 \left[ I_x (\mathbf{x} \mathbf{x} - \mathbf{y} \mathbf{y}) + I_y (\mathbf{x} \mathbf{y} + \mathbf{y} \mathbf{x}) + i l_z (\mathbf{x} \mathbf{y} - \mathbf{y} \mathbf{x}) \right] + \sum_p \left( \frac{f_{sp,pp+q}(\mathbf{p} \mathbf{z}_p - \mathbf{z}_p \mathbf{p}) + f_{sp,qpp}(\mathbf{z}_p \mathbf{p} - \mathbf{p} \mathbf{z}_p)}{E_{c,q+p} + E_{l,p} - i \omega q} - \frac{1}{3} (E_h \rightarrow E_i) \right) + \frac{2}{3} \times
$$

$$
\left[ \frac{f_{sp,pp+q}(\mathbf{p} \mathbf{z}_p - \mathbf{z}_p \mathbf{p}) + f_{sp,qpp}(\mathbf{z}_p \mathbf{p} - \mathbf{p} \mathbf{z}_p)}{E_{c,q+p} + E_{l,p} - i \omega q} \right],
$$

(32)

where $f_{sp,p} \equiv \mathbf{s}_p \cdot \mathbf{e}_p$. Using the anti-symmetric tensor $\mathbf{e} \equiv \mathbf{e}_j \mathbf{e}_j \mathbf{e}_k$ which is invariant under orthogonal coordinate transformation, we can express $\mathbf{x}_p \mathbf{y}_p - \mathbf{y}_p \mathbf{x}_p = \mathbf{z}_p \cdot \mathbf{e} \cdot \mathbf{e}$, $\mathbf{x}_p \mathbf{z}_p - \mathbf{z}_p \mathbf{x}_p = \mathbf{e} \cdot \mathbf{e} \cdot \mathbf{z}_p$, and $\mathbf{z}_p \mathbf{x}_p - \mathbf{x}_p \mathbf{z}_p = \mathbf{y}_p \cdot \mathbf{e}$, whereby the terms associated with the electron spin polarization form an anti-symmetric tensors. Noticing that the contraction between the anti-symmetric and the symmetric tensors associated with $I_i$ and $I_l$ must vanish, and also that the effective Hamiltonian must be real, we have

$$
\mathcal{H}_{\text{eff}}^{HL} = -|\mathbf{d}_c|^2 I_x (\mathbf{x} \mathbf{y} - \mathbf{y} \mathbf{x}) \sum_p \left[ \frac{\mathbf{s}_{p+q} \cdot \mathbf{z}_p \mathbf{z} \cdot \mathbf{e}}{E_{c,q+p} + E_{l,p} - i \omega q} - \frac{\mathbf{s}_{p+q} \cdot \mathbf{z}_p \mathbf{z} \cdot \mathbf{e}}{E_{c,q+p} + E_{l,p} - i \omega q} + \frac{2}{3} \frac{\mathbf{s}_{p+q} \cdot \mathbf{z}}{E_{c,q+p} + E_{l,p} - i \omega q} \right]
$$

(33a)

$$
= 2 |\mathbf{d}_c|^2 I_x \left[ \left( \frac{\mathbf{s}_{p+q} \cdot \mathbf{z}_p \mathbf{z}}{E_{c,q+p} + E_{l,p} - i \omega q} - (E_h \rightarrow E_i) \right) + \frac{2}{3} \frac{\mathbf{s}_{p+q} \cdot \mathbf{z}}{E_{c,q+p} + E_{l,p} - i \omega q} \right].
$$

(33b)

By expanding to the first order of $\mathbf{q}$, we have $E_{c,q+p} \approx E_{c,p} + \mathbf{q} \cdot \mathbf{p}_{c,p}$, and $s_{p+q} \approx s_p + \mathbf{q} \cdot \mathbf{p}_{s,p}$. By using $\nabla_p \mathbf{z}_p = \nabla_p (\mathbf{p}/p) = f^3/p - pp/p^3 = (\mathbf{x}_p \mathbf{x}_p + \mathbf{y}_p \mathbf{y}_p)/p$, and $\nabla_p (\mathbf{z}_p \mathbf{p}) = (\mathbf{x}_p \mathbf{z}_p \mathbf{x}_p + \mathbf{y}_p \mathbf{z}_p \mathbf{y}_p + \mathbf{x}_p \mathbf{z}_p \mathbf{y}_p + \mathbf{y}_p \mathbf{z}_p \mathbf{x}_p)/p$, we obtain the effective Hamiltonian
where \( E_F \) is the Fermi energy, \( \Delta_{hi} \) is the light detuning from the HH/LH band to the Fermi level, respectively [see Fig. 1(a)]. The first term in Eq. (34) results from the spin polarization, while the other terms result from a spin current. When neglecting the HH-LH splitting by letting \( \Delta_h = \Delta_l \) and \( m_h = m_l \), Eq. (34) is reduced to a expression similar to Eq. (30) but with a minus sign \( (m_r \rightarrow m_h, \Delta_r \rightarrow \Delta_h) \). This reduction confirms that the effective Hamiltonian from the HH/LH-CB transitions can be derived as easy as that from the SO-CB transitions if the spin quantization direction in HH/LH band can be chosen arbitrarily. Moreover, if there were no spin-orbit coupling in the valence bands, i.e., the HH, LH, and SO bands had the same effective mass and the same band-edge energy, the coupling between a spin current and a light would vanish.

Finally, once the spin distribution is specifically given, the total effective Hamiltonian will be determined. We assume that the electron spin distribution around Fermi wavevector \( k_F \) deviate only slightly from the equilibrium distribution. More specifically, we suppose the spin distribution has the form

\[
s_p = N_0 + N_1 f(p) \cos \theta_p,
\]

where \( \theta_p \) is the angle between the momentum \( p \) and the current direction \( \mathbf{Z} \). Such a distribution is the usual case for weak currents. A straightforward integration over the momentum space gives [see Appendix A]

\[
\mathcal{H}_\text{eff}^0 = \zeta_0 l_z \mathbf{z} \cdot \mathbf{S},
\]

\[
\mathcal{H}_\text{eff}^1 = \zeta_1 l_z l_z + \zeta_2 q l_z \mathbf{z} \cdot \mathbf{z},
\]

with the coupling constants

\[
\zeta_0 \equiv 2 |d_{cv}|^2 \left( \frac{1}{\Delta_h} + \frac{1}{\Delta_l} - \frac{2}{\Delta_r} \right),
\]

\[
\zeta_1 \equiv \frac{|d_{cv}|^2}{e} \left( \frac{2m_e}{5m_h\Delta_h^2} - \frac{2m_e}{5m_l\Delta_l^2} - \frac{3}{5\Delta_h E_F} + \frac{3}{5\Delta_l E_F} \right),
\]

\[
\zeta_2 \equiv \frac{|d_{cv}|^2}{e} \left( \frac{4m_e}{5m_h\Delta_h^2} + \frac{8m_e}{15m_l\Delta_l^2} - \frac{4m_e}{3m_h\Delta_h^2} - \frac{1}{5\Delta_h E_F} + \frac{1}{5\Delta_l E_F} \right).
\]

For a spin distribution different from Eq. (35), as can be seen in Sec. IV D, the coupling constants shown above will only be changed quantitatively, which further confirms the symmetry analysis in Sec. II [see Eq. (9)].

C. Second-order nonlinear optical effects

The linear optical effect of spin currents is weak since the photon current involves the small light momentum. If we replace the light momentum by another optical field, the coupling can be greatly enhanced by a factor of \( \mathbf{F}_2 \cdot \mathbf{v} / q \cdot \mathbf{v} \). As shown in Sec. II B the second-order nonlinear optical effects of spin currents is rooted in their unique physical nature and spatial inversion-symmetry breaking. Specially, noticing that a longitudinal spin current, is a chiral quantity, we envisaged that it could be probed by the chiral sum-frequency optical (SFG) spectroscopy which was recently developed to detect molecular chirality\(^{13,16}\) If otherwise measured in linear optics, the effect of the chirality relies on the small magnetic moment of the molecules, and in turn the small wave vector of the probe light, similar to the case of linear optical effects of spin currents\(^{20}\).

1. Physical picture

The nonlinear coupling between a spin current and light contains three processes: one virtual interband transition creating an electron-hole pair, one intraband transition accelerating the electron or the hole, and one virtual transition inducing the combination. The physical picture for the microscopic mechanism of the second-order nonlinear optical effects of spin currents is similar to the linear optical effect. A spin will induce a Faraday rotation \( \mathbf{P}^{(1)} \propto \mathbf{F} \times \mathbf{s}_k / (\omega - E_k) \).

\[\begin{aligned}
\mathbf{F}_1 \quad \mathbf{F}_2 \\
\mathbf{s}_k @ -\mathbf{k} \quad \mathbf{s}_k @ \mathbf{k}
\end{aligned}\]

FIG. 3: (color online) Physical picture for the microscopic mechanism of the second-order nonlinear optical effects of a pure spin current. The second light \( \mathbf{F}_2 \) will accelerate the electrons (or holes).
The Faraday rotations due to the pair of spins of \( s_k \) (at momentum \( k \)) and \( -s_k \) (at momentum \( -k \)) cancel each other in the vertical optical transition. Instead of considering the small light-momentum in the linear optical effect, we add another optical field \( F_2 \). The spin will experience an intraband acceleration by this optical field and the transition energy will be changed to \( E_{\pm} \rightarrow E_{\pm} \pm \int e\mathbf{v}_k \cdot F_2 e^{-i\omega t} dt \) [Fig. 3]. The physical meaning of \( e\mathbf{v}_k \cdot F_2 \) is that the power is done by the field to the electron. Therefore, \( \pm i\mathbf{k} \) will induce different Faraday rotation due to opposite energy modification

\[
P^{(2)} \propto \mathbf{F}_1 \times s_k e\mathbf{v}_k \cdot F_2 / ((\omega_1 + \omega_2 - E_k)(\omega_1 - E_k)\omega_2).
\]

This gives the second-order nonlinear optical effects of spin currents.

2. Microscopic calculation

The second-order nonlinear susceptibility can be obtained straightforwardly through the standard perturbation method as shown below. Here we take the SFG as an example of the second-order nonlinear optical effects of spin currents.

The dipole density operator for the intraband transition reads[21]

\[
\hat{P}_\text{intra}(r) = \imath e \sum_{\mu, k} \sum_{\mu', \mathbf{p}} \hat{e}_\mu' \mathbf{p} \cdot \hat{e}_\mu \cdot \langle \mu | p \mu \rangle \mathbf{k}_t + \sum_{j, m, j', m'} \hat{V}_{j, m', j}' \hat{V}_{j, m, k}(j, m' | p, j, m) \rangle \mathbf{k}_t e^{i\mathbf{p} \cdot \mathbf{r} - k \cdot \mathbf{r}}.
\]

With the input optical field consisting of several frequency components \( \mathbf{F}(r, t) = \sum_{j=1,2} F_j e^{-i\omega_j t} + \text{c.c.} \), the light-matter interaction Hamiltonian is

\[
\hat{H}_1(t) = - \left( \hat{D} + \hat{D}^\dagger \right) \cdot \left( \sum_{j=1,2} F_j e^{-i\omega_j t} + \text{c.c.} \right).
\]

with

\[
\hat{D} = - d_v \sum_{\mathbf{k}, \mu} \left( n_{\mu, k} \hat{h}_{\mu, -k} \hat{e}_\mu \cdot \mathbf{k}_t + (1/\sqrt{3}) n_{\mu, k} \hat{h}_{\mu, -k} \hat{e}_\mu \cdot \mathbf{k}_t - \sqrt{2/3} \mathbf{k}_t \hat{\nu}_{\mu, k} \hat{e}_\mu \cdot \mathbf{k}_t + (\mu/\sqrt{3}) \mathbf{k}_t \hat{\nu}_{\mu, k} \hat{e}_\mu \cdot \mathbf{k}_t \right),
\]

\[
\hat{d} = \imath e \sum_{\mu, \mathbf{p} = \pm} \left( \sum_{j, m} \hat{V}_j \hat{\nu}_{j, m} \hat{e}_\mu \cdot \mathbf{k}_t + \sum_{j', m'} \hat{V}_{j', m'} \hat{\nu}_{j', m'} \hat{e}_\mu \cdot \mathbf{k}_t \right) \mathbf{k}_t e^{i\mathbf{p} \cdot \mathbf{r} - k \cdot \mathbf{r}} - \imath e \sum_{\mu, \mathbf{p} = \pm} \left( \sum_{j, m} \hat{V}_j \hat{\nu}_{j, m} \hat{e}_\mu \cdot \mathbf{k}_t + \sum_{j', m'} \hat{V}_{j', m'} \hat{\nu}_{j', m'} \hat{e}_\mu \cdot \mathbf{k}_t \right) \mathbf{k}_t e^{i\mathbf{p} \cdot \mathbf{r} - k \cdot \mathbf{r}}.
\]

denoting the inter- and intra-band polarization operators, respectively. \( \hat{D} \) and \( \hat{D}^\dagger \) are the positive- and negative-frequency components of the inter-band polarization operator, respectively. The first part of the intra-band polarization is the usual acceleration term. The second part, which has the form of non-Abelian Berry connections (similar to vector potentials), accounts for the variation of the spin quantization direction with acceleration of an electron. It is necessary to include the Berry connection term for the gauge-invariance of the inter-band polarization. The explicit form of the Berry connection term depends on the choice of the local coordinate \((\mathbf{x}_p, y_p, z_p)\) at momentum \( \mathbf{p} \). In Appendix B we present an example for the Berry connection in a specific convention.

We adopt the interaction picture for calculating the SFG. The second-order polarization response obtained by the standard perturbation theory is

\[
P^{(2)}(t) = - \int_{-\infty}^{t} dt' \int_{-\infty}^{t} dt'' e^{-i\omega t' - i\omega t''} \text{Tr} \left( \hat{D}(t) \mathbf{F}_2 \cdot \hat{D}^\dagger(t') \left[ \mathbf{F}_1 \cdot \hat{d}(t'), \hat{\rho}_0 \right] + \left[ \mathbf{F}_1, \omega_1 \leftrightarrow \mathbf{F}_2, \omega_2 \right] \right).
\]

where \( \hat{D}(t), \hat{D}_1 \) are operators in the interaction picture. We consider the case that (1) the sum frequency \( \omega = \omega_1 + \omega_2 \) is near resonant with the band-edge, so the positive-frequency component \( \hat{D}(t) \) dominates the optical process; (2) the intra-band dipole moment must be considered for the contribution by the spin current; and (3) no holes exist in the initial system, so the inter-band excitation has to be involved (caused by \( \hat{D}^\dagger \)). With all these considerations taken into account, the second-order response of interest is

\[
P^{(2)}(t) = \int_{-\infty}^{t} dt' \int_{-\infty}^{t} dt'' e^{-i\omega t' - i\omega t''} \text{Tr} \left( \hat{D}(t) \mathbf{F}_2 \cdot \hat{D}^\dagger(t') \left[ \mathbf{F}_1 \cdot \hat{d}(t'), \hat{\rho}_0 \right] + \left[ \mathbf{F}_1, \omega_1 \leftrightarrow \mathbf{F}_2, \omega_2 \right] \right).
\]
The physical meaning of Eq. (42) is clear: Eq. (42a) corresponds to the driving of the electron population (at $t''$) followed by inter-band excitation (at $t'$) and emission (at $t$); Eq. (42b) corresponds to the process in which an electron-hole pair (created at $t''$) is driven by an external field (at $t'$) till its emission (at $t$).

When the HH-LH splitting is neglected, we have a simple microscopic calculation as discussed in Ref. 21 in which the spin quantization for valence band states and the selection rule for interband transitions are independent of its momentum. Beyond such an approximation, the calculation of $P^\parallel$ through Eq. (42) is lengthy, but only quantitatively modifies the results. So we will only list the result in the Appendix E, and the details are shown in the Supplementary Information.

IV. DISCUSSIONS AND NUMERICS

A. Faraday rotation of a spin current and spin polarization

The Faraday rotation angle is expressed as
\[ \theta_F = \frac{\omega l (\chi_{++} - \chi_{--})}{(4nc)}, \]
where $l$ is the light propagation distance, $n$ is the material refractive index, and $c$ is the light velocity in vacuum [Appendix C].

Pure spin current. For a spin current configuration as shown in Fig. 4 where a light comes in with a zenith angle $\beta$ and an azimuth angle $\gamma$, the Faraday rotation angle due to different components of $\mathbf{JZ}$ is
\[
\begin{align*}
\theta_F^{(1)}(J_X) &= \delta_F^{(1)} J_X \xi_2 \sin \beta \cos \gamma, \\
\theta_F^{(1)}(J_Y) &= -\delta_F^{(1)} J_Y \xi_1 \sin^2 \beta \sin \gamma \cos \gamma (n^2 - \sin^2 \beta)^{-1/2}, \\
\theta_F^{(1)}(J_Z) &= \delta_F^{(1)} J_Z (\xi_1 n^2 + \xi_2 \sin^2 \beta \cos^2 \gamma / n) (n^2 - \sin^2 \beta)^{-1/2},
\end{align*}
\]
where $\delta_F^{(1)} = \pi l / 2n_0 \epsilon_0 c^2$. The dependence of the rotation angle on the incident angles for $J_Z$, $J_Y$ and $J_X$ components of a pure spin current are shown in turn in Fig. 5(a), (b) and (c).

Net spin polarization. The net spin polarization also causes the Faraday rotation. With the incident light of zenith angle $\beta$, the Faraday rotation angle equals
\[ \theta_F^{(0)}(\mathbf{S}) = (2\pi l / 8e_0 n_\lambda) (\xi_0 \mathbf{z} \cdot \mathbf{S}). \]
Spin polarization has both the normal and parallel components with respect to the sample surface $\mathbf{S} = \mathbf{S}_\perp + \mathbf{S}_\parallel$. For the normal component $\mathbf{S}_\perp$, the rotation is independent of $\beta$,
\[ \theta_F^{(0)}(\mathbf{S}_\perp) = \pi \xi_0 S_\perp L / 4e_0 n_\lambda, \]
while for parallel component $\mathbf{S}_\parallel$,
\[ \theta_F^{(0)}(\mathbf{S}_\parallel) = (\pi \xi_0 S_\parallel L / 4e_0 n_\lambda) \sin \beta \cos \gamma (n^2 - \sin^2 \beta)^{-1/2}. \]

In general, the angle dependence of Faraday rotation can be used to distinguish a pure spin current from a spin polarization. However, in many materials $n \gg 1 \geq \sin \beta$, both $\theta_F^{(0)}(\mathbf{S})$ and $\theta_F^{(1)}(\mathbf{S})$ have nearly the same angle dependence, which is proportional to $\sin \beta \cos \gamma$. As there is inversion symmetry difference between a pure spin current ($P = -$, odd) and a spin polarization ($P = +$, even), a pure spin current would have a sign flip at reflection while a spin polarization would not. Therefore, the Faraday rotation angle of a pure spin current vanishes through reflection, while the rotation angle of a spin polarization will be doubled. This difference can be used.
to distinguish the effect of a spin current from that of spin polarization.

For the realistic case in Ref. [8] the vanishing Faraday signal is reported in the middle region where the spin current flows without net spin polarization. We explain it with the fact that in the experiment \( Z \rightarrow 0 \) and \( J_z \rightarrow 0 \). With the experimental configuration shown in Fig. 4, the rotation angle \( \gamma \) is reached when \( \beta \rightarrow \pi/2 \) and \( \gamma \rightarrow 0 \). The dependence of maximum Faraday rotation angle on the light wavelength is plotted in Fig. [5d]. For the specific example shown in Fig. [5d] with light wavelength around 800 nm, the maximum Faraday rotation angle is 0.38 \( \mu \)rad. Such a Faraday rotation angle, though still small, is measurable in experiments.

B. Effects of valence band anisotropy

In the derivation above, we have neglected the anisotropy of the valence bands. Now we examine the effect of the valence bands anisotropy. The anisotropic valence band Hamiltonian takes the form

\[
H_{KK}^A = \frac{1}{2m_0} \left[ (\gamma_1 + 5\gamma_2/2) \nabla^2 - 2\gamma_3 (\nabla \cdot \mathbf{K})^2 + 2(\gamma_3 - 2\gamma_1) (\nabla^2 K^2_c + c. p.) \right],
\]

where the \( (\gamma_3 - 2\gamma_1) \) term describes the anisotropy. The anisotropy is usually small. The eigenfunctions of \( H_{KK}^A \) are

\[
|\psi_i\rangle = \sum_{j=\pm 1/2, \pm 1} \alpha_j^i |3/2, j\rangle, \quad i = \pm 3, \pm 1,
\]

where the basis states \( |3/2, \pm 1/2\rangle \) and \( |3/2, \pm 1\rangle \) are explicitly given in Appendix D and \( \alpha_j^i \) are coefficients satisfying \( U^* \alpha = \alpha' \), with \( U = -i\sigma_z \otimes \sigma_y \). The eigenstates \( |\psi_{\pm 1}\rangle \) and \( |\psi_{\pm 1}\rangle \) have eigenvalues \( E_h \) and \( E_l \), respectively. The dipole density operator can be explicitly written as

\[
\hat{\mathbf{P}}(\mathbf{r}) = -e \sum_{\mu, \mu'} \alpha^i_{\mu, \mu'} e^{i \mathbf{p} \cdot \mathbf{r}} \left[ \hat{L}_{-\mu} \hat{\psi}_{\mu, \mu'} \langle \psi_{i}|\mathbf{r}|\mu\rangle + \hat{\psi}_{-\mu, \mu'} \langle \mu|\mathbf{r}|\psi_{i}\rangle \right]
\]

\[
+ \hat{\psi}_{-\mu, \mu'} \langle \mu|\mathbf{r}|\psi_{i}\rangle + \hat{L}_{-\mu} \hat{\psi}_{\mu, \mu'} \langle \psi_{i}|\mathbf{r}|\mu\rangle \right] + H.c.,
\]

where \( |\mu\rangle = |\pm\rangle \) denotes the CB electron state with spin \( \pm 1/2 \), and the operators \( \hat{L}_z \) and \( \hat{\psi}_z \) annihilate \( |\psi_{\pm 1}\rangle \) and \( |\psi_{\pm 1}\rangle \), respectively. By using the fact \( \mathbf{p}/m_0 = dr/dt = (rH_0 - H_0 dr)/i \), we get

\[
\langle \psi_i| - e\mathbf{r}|\mu\rangle = \sum_{j=\pm 1/2, \pm 1} M_{i, \mu} A^{\dagger}_{j, \mu} |j\rangle,
\]

where \( M_{\pm 3/\pm 1} = -ie/m_0 (E_{\pm 3} - E_{\pm 1}) \). The detailed expression for \( A^{\dagger}_{j, \mu} \) can be found in Appendix C. The effective Hamiltonian then reads

\[
H_{ei}^{eff} = Tr \left\{ \hat{\mathbf{P}} \right\} = \sum_{\mu, \mu', \mu''} F_{\mu, \mu'} F_{\mu', \mu''} \alpha_{\mu, \mu'} \alpha_{\mu', \mu''} \langle \mu, \mu'|j\rangle \langle j|\mu', \mu''\rangle,
\]

\[
\left| M_{i, \mu} \right|^2 A^{\dagger}_{j, \mu} A^{\dagger}_{j, \mu'} |j\rangle \left( 1 - \frac{1}{2} \frac{\omega_{\mu'} + \omega_{\mu} + \omega_{\mu''}}{\omega_{\mu'} + \omega_{\mu''}} \right) + \frac{1}{2} \frac{\omega_{\mu'}}{\omega_{\mu'} + \omega_{\mu''}}
\]

The calculation is lengthy. Here we omit the details but just give the terms with \( i = 3, \mu = \mu' = + \) and \( i = -3, \mu = \mu' = - \) explicitly, which is proportional to

\[
(A_{3+}^1 (r_{xp}^2 p_{yp} + |A_{3+}^1 |^2 p_{zp}^2 z) (f_{++} f_{--}) + 2i |(A_{3+}^1 A_{3-}^1)(x_{yp} y_{p} - y_{yp} x_{p}) + 2i |(A_{3+}^1 A_{3-}^1)(y_{zp} z_{p} - z_{yp} y_{p})
\]

\[
+ 2i |(A_{3+}^1 A_{3-}^1)(z_{zp} x_{p} - x_{zp} z_{p}) | f_{p}. \]

The term \( (A_{3+}^1 A_{3-}^1) \) is just a background. The term \( (A_{3+}^1 A_{3-}^1) \) is the total anisotropy tensor, which couples to \( I_z \) only. This result confirms the symmetry analysis in Sec. II.

C. Second-order nonlinear optical effects

The independent parameters of the susceptibility of spin current in a bulk GaAs in Eqs. (19) and (20) are listed in Appendix F. For the sake of simplicity, we neglected the anisotropy of the valence bands. We also neglected the Coulomb interaction, since it is largely screened in the n-doped material. These approximations, according to the symmetry analysis, would only quantitatively modify the results. The bulk inversion asymmetry would cause a background second-order susceptibility, which is indeed strong but can be well separated from the spin-current effect by ac modulation of the current and the phase-locking detection technique. Two representative results of the calculated susceptibility spectra are shown in Fig. 6. The other terms of the susceptibility tensor (not shown) have similar frequency dependence and comparable amplitudes. As a specific example, a transverse spin current 20mA/\( \mu \)m\( ^2 \) has a susceptibility \( \chi^{(2)} \approx 4.8 \times 10^{-12} \text{esu} \) (or \( 0.2 \times 10^{-14} \text{mV} \) in SI units) for input frequencies \( \omega_1 = 100 \text{ meV} \) and \( \omega_2 = 1400 \text{ meV} \) or \( 0.25 \times 10^{-12} \text{esu} \) for \( \omega_1 = \omega_2 = 750 \text{ meV} \) (corresponding to the second harmonics generation).

The SFG of spin current can be straightforwardly extended to other second-order optical spectroscopy such as difference-frequency and three-wave mixing.

V. CONCLUSIONS

In summary, with the systematic symmetry analysis in general and the microscopic calculation under realistic conditions, we have shown that a pure spin current has a measurable circular birefringence effect and a sizable sum-frequency susceptibility. With universality of the method guaranteed by the symmetry principle and without requirements of special structure design and fabrication, the linear and nonlinear optical spectroscopy can be applied to study a wide range of spin-related quantum phenomena such as in topological insulators. A wealth of physics connecting spins and photons and technologies synthesizing spintronics and photonics may be explored.
FIG. 6: Representative results of the sum frequency susceptibility. (a) $-\chi^{(2)}_{YZX}$ due to a longitudinal spin current, and (b) $-\chi^{(2)}_{YYY}$ due to a transverse spin current, as functions of the optical frequencies. Parameters are chosen similar to those in Ref. 8 (same as in Fig. 5). The dielectric constant $\varepsilon = 10.6$, and the spin current $J_X = J_Z = 20 \text{ nA/}\mu\text{m}^2$.

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Appendix A: Coordinate basis

We choose a global coordinate system $(X, Y, Z)$ and define the local coordinates as

\begin{align*}
\hat{\theta}_p &= x_p = X \cos \theta_p \cos \phi_p + Y \cos \theta_p \sin \phi_p - Z \sin \theta_p, \quad (A1a) \\
\hat{\phi}_p &= y_p = -X \sin \phi_p + Y \cos \phi_p, \quad (A1b) \\
\hat{p} &= z_p = X \sin \theta_p \cos \phi_p + Y \sin \theta_p \sin \phi_p + Z \cos \theta_p. \quad (A1c)
\end{align*}

The angle average of the tensor

$$\langle z_p z_p d\Omega \rangle = \frac{1}{4\pi} \int z_p z_p d\Omega = \frac{1}{3} I^{(2)}_Z.$$  \quad (A2)

And the angle average

$$Z \cdot z_p z_p z_p = \frac{1}{4\pi} \int Z \cdot z_p z_p z_p d\Omega = \frac{1}{15} (I^{(2)}_Z + XZX + YZY + ZZZ + ZI^{(2)}).$$ \quad (A3)

For a spin distribution of Eq. 35, the total spin polarization and the spin current is respectively as

$$S = \sum_p s_p = \sum_p N_0,$$  \quad (A4a)

$$J = \sum_p J_p = \frac{e}{m_e} \sum_p N_1 f(p) p Z \cdot z_p z_p,$$

$$= \frac{e}{m_e} \sum_p N_1 f(p) p Z \cdot \bar{z}_p.$$

$$= \frac{N_1 Z}{3} \frac{e}{m_e} \sum_p f(p) p = J_Z.$$ \quad (A4b)

Also, we have

$$\sum_p z_p z_p \cdot \bar{p} = \frac{e}{m_e} \sum_p z_p z_p \cdot (N_1 f(p) p Z \cdot z_p z_p)$$

$$= \frac{e}{m_e} \sum_p f(p) p Z \cdot z_p z_p \cdot N_1$$

$$= \frac{1}{3} \frac{e}{m_e} \sum_p f(p)$$

$$\times \left( I^{(2)}_Z + XZX + YZY + ZZZ + ZI^{(2)} \right) \cdot N_1$$

$$= \frac{1}{5} \left( J_Z I^{(2)} + \bar{J} + \bar{J}^T \right).$$ \quad (A5)

Appendix B: Berry connection

The band edge state of CB are

$$|+/\rangle_p = |S\rangle \otimes |\uparrow / \downarrow\rangle_p,$$ \quad (B1)

with $|S\rangle$ being a periodic s-wave orbital wavefunction which is isotropic in a unit cell, and $|\uparrow / \downarrow\rangle_p$ denoting the spin eigen state parallel/anti-parallel to the momentum.
Similarly, the band edge states of the valence bands are

\[ \begin{align*}
\left[ \frac{3}{2} + \frac{1}{2} \right]_p = & \frac{|X_p + iY_p|}{\sqrt{2}} \otimes |\uparrow_p, \\
\left[ \frac{3}{2} - \frac{1}{2} \right]_p = & \frac{|X_p - iY_p|}{\sqrt{2}} \otimes |\downarrow_p. \\
\end{align*} \] (B2a)

Then the energy density in the material is

\[ \rho_E = \frac{1}{2} \langle \mathbf{D}(r, t) \cdot \mathbf{E}(r, t) \rangle + \frac{1}{2} \langle \mathbf{B}(r, t) \cdot \mathbf{H}(r, t) \rangle = (\varepsilon_0 \varepsilon_r \mathbf{F} + \mathbf{P}) \cdot \mathbf{F}^* + \text{c.c.}, \] (C2)

where \( \varepsilon_r \) is the background dielectric constant. Thus the linear optical susceptibility is related to the effective Hamiltonian through

\[ \mathcal{H}_{\text{eff}} = \varepsilon_0 \sum_{\sigma, \sigma'} \chi_{\sigma, \sigma'} F_{\sigma}^* F_{\sigma'} + \varepsilon_0 \sum_{\sigma, \sigma'} \chi_{\sigma, \sigma'}^* F_{\sigma} F_{\sigma'}^*. \] (C3)

Thus we have

\[ \chi_{\sigma, \sigma'} + \chi_{\sigma, \sigma'}^* = \frac{1}{\varepsilon_0} \frac{\partial^2 \mathcal{H}_{\text{eff}}}{\partial F_{\sigma} \partial F_{\sigma'}^*}. \] (C4)

The index change due to two circular polarization is respectively

\[ \delta n_\pm = \sqrt{\varepsilon_+ - \varepsilon_\pm} - \sqrt{\varepsilon_+} \approx \pm \frac{1}{2} \delta n_\pm \simeq \pm \frac{1}{2} n^{-1} \chi_{\pm \pm}, \] (C5)

where \( n \) is the material refractive index. The phase delay within a propagation length \( l \) is then

\[ \delta \phi_\pm = \omega_0 c^{-1} \delta n_\pm = 2 \lambda_l^{-1} \delta n_\pm, \] (C6)

where \( \lambda_l \) is the light wavelength in vacuum. For a light with linear polarization

\[ \mathbf{x} = (-\mathbf{n}_+ + \mathbf{n}_-) / \sqrt{2}, \] (C7)

after propagation of the length \( l \), the polarization becomes

\[ (\mathbf{n}_+ e^{-i \phi_+} + \mathbf{n}_- e^{-i \phi_-}) / \sqrt{2} = \cos \delta \phi_\pm \mathbf{x} + \sin \delta \phi_\pm \mathbf{y}. \] (C8)

So the Faraday rotation angle is

\[ \theta_F = \delta \phi_+ = \frac{2\pi l}{\lambda} \chi_{\pm \pm}. \] (C9)

For a light with incident zenith and azimuth angles \( \beta \) and \( \gamma \), the angles inside the sample \( \beta' \) and \( \gamma' \) are determined by

\[ \begin{align*}
\sin \beta' &= \sin \beta, \\
\gamma' &= \gamma. \\
\end{align*} \] (C10a)

(C10b)

The propagation length through a sample of thickness \( L \) is

\[ l = L / \cos \beta'. \] (C11)

For a pure spin current and an off-resonant probe, the susceptibility is

\[ \chi_{++}^{(1)} = -\chi_{--}^{(1)} = \frac{1}{4 \varepsilon_0} (\zeta_1 q \mathbf{J}_Z + \zeta_2 q \mathbf{J} \cdot \mathbf{J} \cdot \mathbf{Z}), \] (C12)

Thus the Faraday rotation for a spin current polarized normal to the surface (as in Awschalom’s experiment) is

\[ \theta_F^{(1)} = \delta \phi_+ = \frac{2\pi q L}{8 \varepsilon_0 \varepsilon_r \lambda \cos \beta'} \zeta_1 J \cos \beta' \sin \beta' \cos \gamma \]
\[ \frac{\pi \zeta_2 J L}{2 \varepsilon_0 \varepsilon_r \lambda} \sin \beta', \] (C13)

where \( q = 2\pi n / \lambda \) has been used.

Appendix C: Faraday rotation angle

For a light with frequency \( \omega_0 \), the polarization density is

\[ \mathbf{P} = \varepsilon_0 \sum_{\sigma, \sigma'} \mathbf{n}_\sigma \chi_{\sigma, \sigma'} F_{\sigma'}. \] (C1)

\[ \text{Appendix C: Faraday rotation angle} \]

\[ \text{For a light with frequency } \omega_0, \text{ the polarization density is} \]
\[ \mathbf{P} = \varepsilon_0 \sum_{\sigma, \sigma'} \mathbf{n}_\sigma \chi_{\sigma, \sigma'} F_{\sigma'}. \] (C1)
Appendix D: Anisotropic valence band effect

The anisotropic Luttinger-Kohn matrix of $H_{L_K}^A$ is

$$H_{L_K}^A = \begin{pmatrix} E_3 & P & Q & 0 \\ P^* & E_1 & 0 & Q \\ Q^* & 0 & E_{-1} & -P \\ 0 & Q^* & -P^* & E_{-3} \end{pmatrix},$$  \hspace{1cm} (D1)

where

$$E_3 = E_{-3} = \frac{1}{2m_0} \left[ (\gamma_1 + \gamma_2)k_x^2 - 3\gamma_2k_y^2 \right],$$  \hspace{1cm} (D2a)

$$E_1 = E_{-1} = \frac{1}{2m_0} \left[ (\gamma_1 - \gamma_2)k_x^2 + 3\gamma_2k_y^2 \right],$$  \hspace{1cm} (D2b)

$$P = -\frac{\sqrt{3}y_3}{m_0}k_z(k_x - ik_y),$$  \hspace{1cm} (D2c)

$$Q = \frac{1}{2m_0} \left[ -\sqrt{3}y_2(k_y^2 - k_z^2) + i2\sqrt{3}y_3k_xk_y \right].$$  \hspace{1cm} (D2d)

The eigenstates can be in general written as

$$|\psi_i\rangle = \sum_{j=\pm\pm\pm\pm} \alpha_j^3 |j\rangle, \hspace{0.5cm} i = \pm 3, \pm 1.$$  \hspace{1cm} (D3)

By making the transformation $U^\dagger HU^\dagger \alpha = U^\dagger \alpha$ with

$$U^\dagger = \begin{pmatrix} 0 & 0 & 0 & -1 \\ 0 & 0 & 1 & 0 \\ 0 & -1 & 0 & 0 \\ 1 & 0 & 0 & 0 \end{pmatrix},$$  \hspace{1cm} (D4)

we can see $U^\dagger HU = H^*$. Thus we get the relation

$$U^\dagger \alpha = \alpha'. \hspace{1cm} (D5)$$

Without loss of generality, we take the coordinate relation between the electron and the crystal as

$$(010) = \sin \tilde{\phi} x_p - \cos \tilde{\phi} y_p$$

$$(010) = \cos \tilde{\theta} \cos \tilde{\phi} x_p + \cos \tilde{\theta} \sin \tilde{\phi} y_p - \sin \tilde{\theta} z_p$$

$$(001) = \sin \tilde{\theta} \cos \tilde{\phi} x_p + \sin \tilde{\theta} \sin \tilde{\phi} y_p + \cos \tilde{\theta} z_p \hspace{1cm} (D6)$$

where (001), (010) and (100) are directions of the three crystal axis, and $\tilde{\theta}$ and $\tilde{\phi}$ are the relative direction angles between $x_p, y_p, z_p$ and the (100), (010), (001) axes.

The explicit form of $A_{i,\mu}^j$ is

$$A_{i,\mu}^x = \pm \frac{1}{\sqrt{2}} \left( \alpha_{i}^{\pm\dagger} \right)^* \left[ \sin \tilde{\phi} + i \left( \frac{1}{\sqrt{2}} \left( \alpha_{i}^{\pm\dagger} \right)^* + \frac{1}{\sqrt{6}} \left( \alpha_{i}^{\pm\dagger} \right)^* \right) \cos \tilde{\theta} \cos \tilde{\phi} + \sqrt{\frac{2}{3}} \left( \alpha_{i}^{\pm\dagger} \right)^* \sin \tilde{\theta} \cos \tilde{\phi} \right],$$  \hspace{1cm} (D7a)

$$A_{i,\mu}^y = \pm \frac{1}{\sqrt{2}} \left( \alpha_{i}^{\pm\dagger} \right)^* \left[ \cos \tilde{\phi} + i \left( \frac{1}{\sqrt{2}} \left( \alpha_{i}^{\pm\dagger} \right)^* + \frac{1}{\sqrt{6}} \left( \alpha_{i}^{\pm\dagger} \right)^* \right) \cos \tilde{\theta} \sin \tilde{\phi} + \sqrt{\frac{2}{3}} \left( \alpha_{i}^{\pm\dagger} \right)^* \sin \tilde{\theta} \cos \tilde{\phi} \right],$$  \hspace{1cm} (D7b)

$$A_{i,\mu}^z = -i \left( \frac{1}{\sqrt{2}} \left( \alpha_{i}^{\pm\dagger} \right)^* + \frac{1}{\sqrt{6}} \left( \alpha_{i}^{\pm\dagger} \right)^* \right) \sin \tilde{\theta} + \frac{2}{3} \left( \alpha_{i}^{\pm\dagger} \right)^* \cos \tilde{\theta},$$  \hspace{1cm} (D7c)

and with Eq. (D5), they satisfy the relation

$$A_{i,\mu}^j = -vA_{i,\mu}^{j*} \hspace{1cm} (D8)$$
Appendix E: Second-order nonlinear susceptibility

With a spin current of the form $J = J_X XZ + J_Z ZZ$, where $J_X$ is the transverse amplitude, the second-order nonlinear optical susceptibility induced by the spin current is

$$
\chi^{(2)}(\omega_1, \omega_2; \omega_1 + \omega_2) = \left[J_X \right]^{XY}(2\xi_3 - 2\xi'_3 + \xi'_4 + \xi'_5) + \left[ZZ \right](4\xi_3 - \xi'_3 + \xi'_4 - \xi'_5) + \left[YX \right](4\xi_3 + \xi'_3 - \xi'_4 - \xi'_5) + \left[YZ \right](\xi_3 - \xi'_3 + \xi'_4 - \xi'_5) + \left[YY \right](4\xi_3 - 4\xi'_3),
$$

(E1a)

$$
+ J_Z \left[XYZ - YXZ \right](\xi_3 + \xi'_3 + 3\xi_5 - \xi'_5) + \left[(ZX - YXY \right]2\xi_3 - \xi'_3 - 2\xi'_5 - \xi'_5) + \left[(ZXY - YXY \right]2\xi_3 + 3\xi_5 - \xi'_5 - 3\xi'_5) \right],
$$

(E1b)

where $\xi'_3$ is derived from $\xi_3$ by exchanging $\omega_1$ and $\omega_2$, and

$$
\xi_1 = \left(\frac{e_i + 2}{3}\right)^3 [d_{cv}]^2 \left[\frac{1 + m_s/m_I}{(\Delta^2)^2 \omega_1} + \frac{1 + m_s/m_I}{(\Delta^2)^2 \omega_1} - \frac{1 + m_s/m_I}{(\Delta^2)^2 \omega_1} - \frac{1 + m_s/m_I}{(\Delta^2)^2 \omega_1} \right].
$$

(E2a)

$$
\xi_2 = \left(\frac{e_i + 2}{3}\right)^3 [d_{cv}]^2 \left[\frac{\Delta' - \Delta^h}{2E_F \Delta^h \Delta^h} + \frac{\Delta' - \Delta^h}{2E_F \Delta^h \Delta^h} \right],
$$

(E2b)

$$
\xi_3 = \left(\frac{e_i + 2}{3}\right)^3 [d_{cv}]^2 \left[\frac{1}{5} \left(\Delta' - \Delta^h \right) \left(\Delta'_2 - \Delta^h \right) \right],
$$

(E2c)

$$
\xi_4 = \left(\frac{e_i + 2}{3}\right)^3 [d_{cv}]^2 \left[\frac{\Delta' - \Delta^h}{2E_F \Delta^h \Delta^h} + \frac{\Delta' - \Delta^h}{2E_F \Delta^h \Delta^h} \right],
$$

(E2d)

$$
\xi_5 = \left(\frac{e_i + 2}{3}\right)^3 [d_{cv}]^2 \left[\frac{1 + m_s/m_h}{(\Delta^h)^2 \omega_1} + \frac{1 + m_s/m_h}{(\Delta^h)^2 \omega_1} - \frac{1 + m_s/m_h}{(\Delta^h)^2 \omega_1} - \frac{1 + m_s/m_h}{(\Delta^h)^2 \omega_1} \right].
$$

(E2e)

where the factor containing the material dielectric constant $\epsilon$, takes into account the difference between the macroscopic external field and the microscopic local field.\(^{33}\)
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