

Optical effects of spin currents in semiconductors

Jing Wang,^{1,2,*} Sheng-Nan Ji,^{1,2} Bang-Fen Zhu,^{1,3,†} and Ren-Bao Liu^{2,4,‡}

¹State Key Laboratory of Low-Dimensional Quantum Physics, and Department of Physics, Tsinghua University, Beijing 100084, China

²Department of Physics, The Chinese University of Hong Kong, Shatin, N.T., Hong Kong, China

³Institute of Advanced Study, Tsinghua University, Beijing 100084, China

⁴Institute of Theoretical Physics, The Chinese University of Hong Kong, Shatin, N.T., Hong Kong, China

(Received 11 May 2012; published 24 July 2012)

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 neither 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.

DOI: [10.1103/PhysRevB.86.045215](https://doi.org/10.1103/PhysRevB.86.045215)

PACS number(s): 72.25.Dc, 78.20.Ls, 42.65.An, 78.30.Fs

I. INTRODUCTION

A pure spin current consists of flows of opposite spins in opposite directions with the same amplitude. It bears neither net charge current nor net spin polarization. Spin currents are a key element in spintronics.^{1,2}

Detection of spin currents is important for characterization and applications in future spintronics technologies.^{1,2} While a polarized spin current may be detected by the conventional Faraday/Kerr rotation spectroscopy³⁻⁵ or through ferromagnetic filters,^{6,7} 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,^{8,9} polarized light emission,¹⁰ and polarization-selective absorption.^{11,12} Or the inverse spin Hall effect¹³⁻¹⁶ can be used to convert a spin current into charge/voltage signals for electric measurement.¹⁷⁻²¹ All such measurements, however, disturb the spin currents to some extent and are indirect. We are motivated to find a nondestructive way to directly measure a pure spin current.^{22,23}

A basic symmetry principle states that whenever there is a current breaking the fundamental symmetries of a system, an interaction may arise between the current and another current of the same symmetry-breaking type so that the fundamental symmetries are retained.^{24,25} A classic example is the Ampère effect and the Ørsted effect where a charge current is coupled to another charge current or a magnet. A straightforward analog suggests that a pure spin current may be coupled to another spin current. Such an idea stimulated the proposal of direct measurement of a pure spin current in a direct-gap semiconductor by a polarized light beam.²² A polarized light beam can be regarded as such a “photon spin current”²⁶ by

mapping the photon polarization into a spin 1/2 in the Jones vector representation²⁷

$$\cos \frac{\theta}{2} e^{i\phi/2} \mathbf{n}_+ + \sin \frac{\theta}{2} e^{-i\phi/2} \mathbf{n}_- \sim |\theta, \phi\rangle, \quad (1)$$

where the right/left circular polarization $\mathbf{n}_{+/-}$ 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 that depends on the light polarization and wave vector. The observable result is a circular birefringence effect that is similar to the Faraday rotation but involves no net magnetization nor time-reversal symmetry breaking. Since Faraday’s discovery in 1845,²⁸ 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.²⁹ 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.²² 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 asymmetries³⁰⁻³² is not a necessity, thus the system can bear the inversion symmetry.

The optical birefringence effect of spin currents²² 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 currents. However, if the velocity of spin currents couples to another optical field,

$$\mathbf{q} \cdot \mathbf{v} \Rightarrow \mathbf{F}_2 \cdot \mathbf{v},$$

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,²³ which was soon verified by experiments.³³

In Refs. 22 and 23, 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 pseudospin 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.^{34,35} 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 (~ 0.01 meV in GaAs with doping density $\sim 10^{16}$ cm⁻³) is much less than the detuning of the light from the interband transitions³⁶ that mediate the effective interaction, so it would only slightly modify the coupling coefficients between spin currents and light. 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. Therefore in this paper, we can neglect the inversion asymmetry in the measurement process even though it could be of vital importance in generating the spin current. Without considering the Dresselhaus and Rashba effects due to inversion asymmetries, we avoid the subtlety in the definition of a spin current.^{37,38} It should point out that the Dresselhaus effect under high energy (well above the gap) excitation and the Rashba effect in low-dimensional structures could induce

important effects in the coupling between the light and the spin current. Such effects need further investigation.

The paper is organized as follows. Section 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. Section 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. Section IV presents the numerical results and discussions of the experiment scheme. Section V concludes this paper.

II. SYMMETRY ANALYSIS

We will particularly consider the time-reversal (\mathcal{T}) and the space-inversion (\mathcal{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 \mathcal{P} symmetry, a spin current can induce the second-order nonlinear optical processes.

A. Linear optical effects

We assume the whole system has the \mathcal{T} and \mathcal{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}_{eff} are

$$\begin{array}{c} \hline \hline \mathcal{T} \quad \mathcal{P} \\ \hline \mathcal{H}_{\text{eff}} \quad + \quad + \\ \hline \hline \end{array}, \quad (2)$$

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

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

$$\mathbb{J} = \sum_{\mathbf{p}} \mathbb{J}_{\mathbf{p}} = e \sum_{\mathbf{p}} \mathbf{s}_{\mathbf{p}} \mathbf{v}_{\mathbf{p}}, \quad (3)$$

where $\mathbf{s}_{\mathbf{p}}$ is the spin polarization and $\mathbf{v}_{\mathbf{p}}$ is the velocity of a particle with wave vector \mathbf{p} , and e is the electron charge. The ‘‘photon spin current’’ tensor for a polarized light beam with electric field $\mathbf{F}(\mathbf{r}, t) = (F_+ \mathbf{n}_+ + F_- \mathbf{n}_-) e^{i\mathbf{q}\cdot\mathbf{r} - i\omega_q t} + \text{c.c.}$ is formulated as

$$\mathbb{I} \equiv \mathbf{I}\mathbf{q} \equiv q(I_x \mathbf{x}\mathbf{z} + I_y \mathbf{y}\mathbf{z} + I_z \mathbf{z}\mathbf{z}), \quad (4a)$$

$$I_j = \frac{1}{2} \sum_{\mu, \nu = \pm} \sigma_{\mu\nu}^j F_{\mu}^* F_{\nu}, \quad (4b)$$

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 $\mathbf{n}_{\pm} \equiv (\mp \mathbf{x} - i\mathbf{y})/\sqrt{2}$, and σ^j ($j = x, y, z$) is the Pauli matrix. For completeness, we also consider the spin polarization of the system

$$\mathbf{S} = \sum_{\mathbf{p}} \mathbf{s}_{\mathbf{p}}. \quad (5)$$

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

	\mathbf{S}	\mathbf{q}	\mathbb{J}	I_x, I_y	I_z
\mathcal{T}	-	-	+	+	-
\mathcal{P}	+	-	-	+	+

(6)

It is worth mentioning here there is no $|0\rangle$ state in the physical spin of photons, and the photon pseudospin I_x and I_y do not break the \mathcal{T} symmetry, for it involves the second-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 $I_z \mathbf{z}$. Thus the effective interaction between a spin polarization and a light beam has the form

$$\mathcal{H}_{\text{eff}}^{(0)} = \zeta_0 I_z \mathbf{S} \cdot \mathbf{z}, \quad (7)$$

with a coupling constant ζ_0 to be determined by the specific microscopic mechanism. Such a coupling corresponds to the conventional Faraday effect in magneto-optics.³⁹ 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 I_x and I_y , the spin polarization should be of an even power. Thus to the leading order, the effective interaction has the form

$$\mathcal{H}_{\text{eff}}^{\text{Voigt}} = \zeta_0^{\text{Voigt}} I_x (\mathbf{S} \cdot \mathbf{x})^2. \quad (8)$$

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 I_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, coupling between the spin current and the photon current $qI_z \mathbf{z}$ is possible. The linear birefringence effect (similar to the Voigt effect in magneto-optics) is absent. Due to the lack of $|0\rangle$ state in the physical spin of photons, I_x and I_y preserve \mathcal{T} symmetry. Therefore there is no linear coupling of \mathbb{J} to $qI_x \mathbf{x}$ and $qI_y \mathbf{y}$.

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

$$\mathcal{H}_{\text{eff}}^{(1)} = \zeta_1 q I_z \text{Tr}(\mathbb{J}) + \zeta_2 q I_z \mathbf{z} \cdot \mathbb{J} \cdot \mathbf{z}, \quad (9)$$

with only two coupling constants ζ_1 and ζ_2 to be determined by the microscopic mechanisms. A possible spherically sym-

metric 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

$$\mathcal{H}_{\text{eff}}^{(1)} = q I_z \mathbf{z} \mathbf{z} : \mathcal{A} : \mathbb{J}, \quad (10)$$

where \mathcal{A} is a parameter tensor determined by the microscopic structure of the material. Since only the light polarization term I_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

$$\mathbb{J} = J_x \mathbf{X} \mathbf{X} + J_y \mathbf{Y} \mathbf{Y} + J_z \mathbf{Z} \mathbf{Z} = \mathbf{J} \mathbf{Z}, \quad (11)$$

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} - \mathbf{Z} plane will impose further constraint on the interaction and significantly simplify the Hamiltonian. Under the reflection, the relevant quantities transform as

	q	I_z	\mathbf{J}_{\parallel}	\mathbf{J}_{\perp}
Reflection with \mathbf{z} - \mathbf{Z} plane	+	-	-	+

(12)

where \mathbf{J}_{\parallel} 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}_{\parallel} would couple with the qI_z . Without loss of generality, let \mathbf{Y} be perpendicular to the \mathbf{z} - \mathbf{Z} plane, the effective Hamiltonian reads

$$\mathcal{H}_{\text{eff}}^{(1)} = A_1 q I_z J_z + A_2 q I_z J_x, \quad (13)$$

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

$$\mathcal{H}_{\text{eff}}^{(1)} = \zeta_1 q I_z J_z + \zeta_2 q I_z \mathbf{z} \cdot \mathbb{J} \cdot \mathbf{z}, \quad (14)$$

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,

$$\chi_{\mu, \nu} + \chi_{\nu, \mu}^* = (1/\epsilon_0) \partial^2 \mathcal{H}_{\text{eff}} / (\partial F_{\mu}^* \partial F_{\nu}), \quad (15)$$

where ϵ_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:

$$\chi_{++}^{(0)} = -\chi_{--}^{(0)} = (1/4\epsilon_0)\zeta_0 \mathbf{z} \cdot \mathbf{S}, \quad (16a)$$

$$\chi_{++}^{(1)} = -\chi_{--}^{(1)} = (q/4\epsilon_0)(\zeta_1 J_Z + \zeta_2 \mathbf{z} \cdot \mathbb{J} \cdot \mathbf{z}). \quad (16b)$$

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. Equation (16a) is nothing but the conventional Faraday rotation in magneto-optics.³⁹ Equation (16b) indicates that a pure spin current would produce a circular birefringence effect. This new effect of a pure spin current may be dubbed “*spin current Faraday effect*”²² because of its similarity to the conventional Faraday rotation due to magnetization, with awareness that a pure spin current, however, bears no net magnetization.

B. Second-order nonlinear optical effects

The second-order nonlinear optical effect such as sum-frequency process is characterized by a second-order nonlinear susceptibility $\chi^{(2)}$ via

$$\mathbf{P}^{(2)}(\omega_1 + \omega_2) = \chi^{(2)} : \mathbf{F}_1(\omega_1)\mathbf{F}_2(\omega_2), \quad (17)$$

where \mathbf{F}_1 and \mathbf{F}_2 are the two optical fields, \mathbf{P} is the induced polarization, and $\chi^{(2)}$ is a rank-3 tensor. Under \mathcal{P} operation, \mathbf{F}_1 , \mathbf{F}_2 and \mathbf{P} reverse the sign, which means $\chi^{(2)}$ is zero if the system has \mathcal{P} -symmetry. A pure spin current breaks the \mathcal{P} symmetry, results in a nonzero $\chi^{(2)}$, and makes the second-order nonlinear optical process possible.

In general, as a rank-3 tensor $\chi^{(2)}$ has 27 independent components:

$$\chi^{(2)} = \chi_{XXX}\mathbf{XXX} + \chi_{XXY}\mathbf{XXY} + \dots + \chi_{ZZZ}\mathbf{ZZZ}. \quad (18)$$

But the symmetry properties of the spin current and the system will impose constraints on $\chi^{(2)}$, reducing the number of independent parameters.⁴⁰ For a longitudinal spin current $J_Z\mathbf{ZZ}$ in which the spin polarization is parallel or antiparallel to the current direction the spin current is reversed under the reflection with respect to the X - Z plane, so that $\chi_{XXX}\mathbf{XXX} + \chi_{XXY}\mathbf{XX}(-\mathbf{Y}) + \dots + \chi_{ZZZ}\mathbf{ZZZ} = -\chi^{(2)}$ and the terms with direction \mathbf{Y} appeared even times (twice or zero times) must vanish. Similarly, the longitudinal spin current is reversed under the reflection with respect to the Y - Z and X - Y planes, so the terms with direction \mathbf{X} or \mathbf{Z} that appear even times must be zero. Moreover, the longitudinal spin current should be invariant under the $\pi/2$ rotation with respect to its current direction, therefore $\chi_{XYZ} = -\chi_{YXZ}$, $\chi_{YZX} = -\chi_{XZY}$, $\chi_{ZXY} = -\chi_{ZYX}$. With all these constraints, the sum-frequency susceptibility induced by a longitudinal spin current $J_Z\mathbf{ZZ}$ can be expressed as

$$\chi_{J_Z}^{(2)} = J_Z [\alpha_1(\mathbf{XYZ} - \mathbf{YXZ}) + \alpha_2(\mathbf{YZX} - \mathbf{XZY}) + \alpha_3(\mathbf{ZXY} - \mathbf{ZYX})], \quad (19)$$

where there are only three independent parameters α_i ($i = 1, 2, 3$). For a transverse spin current $J_X\mathbf{XZ}$, in which the spin polarization is perpendicular to the current direction, the current is reversed under reflection with respect to the X - Z plane, but invariant under reflection with respect to X - Y or Y - Z plane. Then the terms that contain even times of \mathbf{Y} or odd

times of \mathbf{Z} or \mathbf{X} must be zero, so

$$\chi_{J_X}^{(2)} = J_X (x_1\mathbf{XXY} + x_2\mathbf{XXY} + x_3\mathbf{YXX} + z_1\mathbf{ZZY} + z_2\mathbf{ZYZ} + z_3\mathbf{YZZ} + y\mathbf{YYY}), \quad (20)$$

with seven independent parameters to be determined. Similar symmetry analysis can be applied to $J_Y\mathbf{YZ}$. 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.^{41,42}

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.^{40,43} We employ the eight-band model.⁴⁴ We assume that the pure spin current result from a nonequilibrium distribution of electrons in the conduction band. Namely, as shown in Fig. 1(a), a small portion of nonequilibrium electrons with opposite velocities near the Fermi surface have opposite spin polarizations, which is similar to the situation in Ref. 8. The optical interaction includes the interband transitions and the intraband acceleration of electrons and holes. To avoid real absorption of light, the light frequencies are chosen to be below the absorption edge in linear optical effect, and the sum frequency is below the two-photon absorption edge in the second-order nonlinear optical effect.

A. Model

We consider an n -doped direct-gap semiconductor of GaAs as a model material. Since other bands are separated far away in energy, we assume the near-gap optical interactions in GaAs involve mostly the eight bands around the fundamental gap, including the conduction band (CB), the heavy-hole (HH)

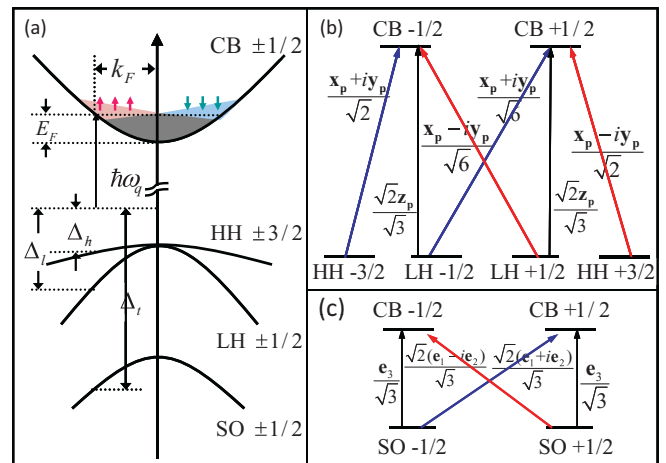


FIG. 1. (Color online) (a) Schematic band structure of the eight-band model near the Γ point of an n -doped III-V compound semiconductor, and illustration of the spin nonequilibrium distribution of electrons for a pure spin current. (b) and (c) Selection rules and relative dipole moments from the HH and LH, SO bands to the CB.

band, the light-hole (LH) band, and the spin-orbit split-off (SO) band, each of two-fold degeneracy [see Fig. 1(a)].

Near the Γ point of the Brillouin zone, the energy dispersion of the CB and the SO electron is almost parabolic and isotropic, which can be respectively written as $E_{ep} = p^2/(2m_e) + E_g$ and $E_{rp} = p^2/(2m_r) + E_{SO}$, where $m_{e/r}$ is the effective mass of the CB/SO band, E_g is the fundamental band gap with the energy zero at the top of the valence band, and E_{SO} is the split-off energy due to the spin-orbit coupling. Hereafter the Planck constant \hbar is taken as unity. For the HH and LH bands, since the energy band anisotropy will not affect the symmetry analysis as shown in Sec. II (the fourfold rotation symmetry will be retained even considering the anisotropic dispersion), we will neglect the anisotropy effect in this section, and take it into account separately in Sec. IV B. Thus we express the isotropic Luttinger-Kohn Hamiltonian H_{LK}^I for the HH and LH bands near the band edge as⁴⁵

$$H_{LK}^I = \frac{1}{2m_0} \left[\left(\gamma_1 + \frac{5}{2}\gamma_2 \right) \nabla^2 - 2\gamma_2 (\nabla \cdot \mathbf{K})^2 \right], \quad (21)$$

where \mathbf{K} is a spin 3/2 for the total angular momentum of an electron in the HH and LH bands, γ_1 and γ_2 are the Luttinger parameters, and m_0 is the free electron mass. The isotropic Luttinger-Kohn Hamiltonian can be diagonalized with the spin 3/2 quantized along the direction of the momentum \mathbf{p} . The HH band with magnetic quantum numbers $\pm 3/2$ has the energy dispersion $E_{hp} = (\gamma_1 - 2\gamma_2)p^2/2m_0 \equiv p^2/2m_h$; and the LH band has magnetic quantum numbers $\pm 1/2$ with the dispersion relation as $E_{lp} = (\gamma_1 + 2\gamma_2)p^2/2m_0 \equiv p^2/2m_l$, where $m_{h/l}$ is the effective mass of the HH/LH band. If the HH-LH splitting is neglected further, the HH and LH bands become a fourfold degenerate spin-3/2 band and the spin quantization direction can be chosen independent of the momentum. The effective mass model for the conduction bands and the Luttinger model for the valence bands do not take into account the Bloch wave function mixing between the conduction and the valence bands, which is justified in this study since only the near band edge excitations are considered. For higher energy excitation, an $8 \times 8 \mathbf{k} \cdot \mathbf{p}$ Kane model would be needed to take into both the mass and wave-function renormalization.

The Bloch state of a CB electron with momentum \mathbf{p} is $|\psi_{\pm}^c(\mathbf{p})\rangle \equiv \hat{e}_{\pm, \mathbf{p}}^{\dagger}|0\rangle = e^{i\mathbf{p}\cdot\mathbf{r}}|\pm\rangle_{\mathbf{p}}$, where $|0\rangle$ represents the vacuum state and $\hat{e}_{\pm, \mathbf{p}}^{\dagger}$ denotes an creation operator that produces an electron in CB with spin $\pm 1/2$ and momentum \mathbf{p} . The Bloch state of an electron in the valence band with momentum \mathbf{p} is $|\psi_m^{\alpha}(\mathbf{p})\rangle \equiv \hat{V}_{j, m, \mathbf{p}}^{\dagger}|0\rangle = e^{i\mathbf{p}\cdot\mathbf{r}}|j, m\rangle_{\mathbf{p}}$, in which $j = 3/2$ and $m = \pm 3/2$ stands for the HH band ($\alpha = h$), $j = 3/2$ and $m = \pm 1/2$ for the LH band ($\alpha = l$), $j = 1/2$ and $m = \pm 1/2$ for the SO band ($\alpha = r$), and $\hat{V}_{j, m, \mathbf{p}}^{\dagger}$ denotes the annihilation operator for an electron in the corresponding valence band. Then the noninteracting Hamiltonian is

$$\hat{H}_0 = \sum_{\mu=\pm, \mathbf{p}} (E_{c\mathbf{p}} \hat{e}_{\mu, \mathbf{p}}^{\dagger} \hat{e}_{\mu, \mathbf{p}} + E_{h\mathbf{p}} \hat{h}_{\mu, \mathbf{p}}^{\dagger} \hat{h}_{\mu, \mathbf{p}} + E_{l\mathbf{p}} \hat{l}_{\mu, \mathbf{p}}^{\dagger} \hat{l}_{\mu, \mathbf{p}} + E_{r\mathbf{p}} \hat{r}_{\mu, \mathbf{p}}^{\dagger} \hat{r}_{\mu, \mathbf{p}}), \quad (22)$$

where the hole operators are defined as $\hat{h}_{\mp, -\mathbf{p}} \equiv \hat{V}_{3/2, \pm 3/2, \mathbf{p}}^{\dagger}$, $\hat{l}_{\mp, -\mathbf{p}} \equiv \hat{V}_{3/2, \pm 1/2, \mathbf{p}}^{\dagger}$, and $\hat{r}_{\mp, -\mathbf{p}} \equiv \hat{V}_{1/2, \pm 1/2, \mathbf{p}}^{\dagger}$. It should be

pointed out that here the angular momentum \mathbf{K} is quantized along \mathbf{p} so that the spin-orbit coupling in the valence bands has already been included.

The initial state of the system (before optical excitation) is characterized by a density matrix $\hat{\rho}_0$. We assume that the system has translation symmetry and initially there is no hole in the system, so we have

$$\text{Tr}[\hat{\rho}_0 \hat{h}_{\mu\mathbf{k}}^{\dagger} \hat{h}_{\nu\mathbf{k}'}] = \text{Tr}[\hat{\rho}_0 \hat{l}_{\mu\mathbf{k}}^{\dagger} \hat{l}_{\nu\mathbf{k}'}] = \text{Tr}[\hat{\rho}_0 \hat{r}_{\mu\mathbf{k}}^{\dagger} \hat{r}_{\nu\mathbf{k}'}] = 0, \quad (23a)$$

$$\text{Tr}[\hat{\rho}_0 \hat{e}_{\mu\mathbf{k}}^{\dagger} \hat{e}_{\nu\mathbf{k}'}] = \delta_{\mathbf{k}, \mathbf{k}'} f_{\mu\nu, \mathbf{k}}. \quad (23b)$$

The spin current, which results from the nonequilibrium 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 $\mathbf{v}_{\mathbf{p}} = \nabla_{\mathbf{p}} E_{c\mathbf{p}}$ and $\mathbf{s}_{\mathbf{p}} = (1/2) \sum_{\mu\nu} \sigma_{\mu\nu} f_{\mu\nu, \mathbf{p}}$ with σ 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 [see Figs. 1(b) and 1(c)], the polarization density operator reads⁴³

$$\hat{\mathbf{P}}_{\text{inter}}(\mathbf{r}) = -d_{cv}^* \sum_{\mathbf{k}, \mathbf{p}; \mu=\pm} \left(\mathbf{n}_{\bar{\mu}, \mathbf{p}} \hat{h}_{\bar{\mu}, -\mathbf{p}} \hat{e}_{\mu, \mathbf{k}} + \frac{1}{\sqrt{3}} \mathbf{n}_{\mu, \mathbf{p}} \hat{l}_{\mu, -\mathbf{p}} \hat{e}_{\mu, \mathbf{k}} - \sqrt{\frac{2}{3}} \mathbf{z}_{\mathbf{p}} \hat{l}_{\bar{\mu}, -\mathbf{p}} \hat{e}_{\mu, \mathbf{k}} - \mu \sqrt{\frac{2}{3}} \mathbf{n}_{\mu, \mathbf{p}} \hat{l}_{\mu, -\mathbf{p}} \hat{e}_{\mu, \mathbf{k}} + \frac{\mu}{\sqrt{3}} \mathbf{z}_{\mathbf{p}} \hat{l}_{\bar{\mu}, -\mathbf{p}} \hat{e}_{\mu, \mathbf{k}} \right) e^{i\mathbf{p}\cdot\mathbf{r} - i\mathbf{k}\cdot\mathbf{r}} + \text{H.c.}, \quad (24)$$

where $\mathbf{n}_{\pm, \mathbf{p}} \equiv \mp(\mathbf{x}_{\mathbf{p}} \pm i\mathbf{y}_{\mathbf{p}})/\sqrt{2}$ denotes the right/left circular polarization about the momentum direction \mathbf{p} , $\mathbf{z}_{\mathbf{p}} \equiv \mathbf{p}/p$, and $\bar{\mu} \equiv -\mu$. As will be discussed in Sec. IV B, 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 $\hat{H}_1(t) = -\int \hat{\mathbf{P}}_{\text{inter}}(\mathbf{r}) \cdot \mathbf{F}(\mathbf{r}, t) d\mathbf{r}$, the light-matter interaction Hamiltonian in the rotating wave approximation can be explicitly expressed as

$$\hat{H}_1 \equiv \exp \left(\sum_{\mu, \mathbf{k}} i\omega_q t \hat{e}_{\mu, \mathbf{k}}^{\dagger} \hat{e}_{\mu, \mathbf{k}} \right) \hat{H}_1(t) \exp \left(\sum_{\mu, \mathbf{k}} -i\omega_q t \hat{e}_{\mu, \mathbf{k}}^{\dagger} \hat{e}_{\mu, \mathbf{k}} \right) = d_{cv}^* \sum_{\mu, \nu, \mathbf{p}} F_{\nu}^* \mathbf{n}_{\nu}^* \cdot \left(\mathbf{n}_{\bar{\mu}, \mathbf{p}} \hat{h}_{\bar{\mu}, -\mathbf{p}} \hat{e}_{\mu, \mathbf{q}+\mathbf{p}} + \frac{1}{\sqrt{3}} \mathbf{n}_{\mu, \mathbf{p}} \hat{l}_{\mu, -\mathbf{p}} \hat{e}_{\mu, \mathbf{q}+\mathbf{p}} - \sqrt{\frac{2}{3}} \mathbf{z}_{\mathbf{p}} \hat{l}_{\bar{\mu}, -\mathbf{p}} \hat{e}_{\mu, \mathbf{q}+\mathbf{p}} - \mu \sqrt{\frac{2}{3}} \mathbf{n}_{\mu, \mathbf{p}} \hat{l}_{\mu, -\mathbf{p}} \hat{e}_{\mu, \mathbf{q}+\mathbf{p}} + \frac{\mu}{\sqrt{3}} \mathbf{z}_{\mathbf{p}} \hat{l}_{\bar{\mu}, -\mathbf{p}} \hat{e}_{\mu, \mathbf{q}+\mathbf{p}} \right) + \text{H.c.} \quad (25)$$

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

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

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,

$$\mathbf{P}^{(1)} \propto \frac{\mathbf{F} \times \mathbf{s}_{\mathbf{k}}}{\omega - E_{\mathbf{k}}}, \quad (27)$$

where $\mathbf{s}_{\mathbf{k}}$ is the spin polarization associated with the state of \mathbf{k} , $E_{\mathbf{k}}$ is the resonant optical transition energy. This naturally explains Faraday rotation due to spin polarization as in Eq. (36a).

For a pure spin current, we first consider only a pair of spins, $\mathbf{s}_{\mathbf{k}}$ at momentum \mathbf{k} , and $-\mathbf{s}_{\mathbf{k}}$ at momentum $-\mathbf{k}$ in the CB [see Fig. 2]. This pair can be viewed as a generator of pure spin current. $\mathbf{s}_{\mathbf{k}}$ gives rise to a Faraday rotation of $\mathbf{P}_{\mathbf{k}}^{(1)} \propto \mathbf{F} \times \mathbf{s}_{\mathbf{k}}/(\omega - E_{+\mathbf{k}})$; while $-\mathbf{s}_{\mathbf{k}}$ leads to a Faraday rotation of $\mathbf{P}_{-\mathbf{k}}^{(1)} \propto \mathbf{F} \times \mathbf{s}_{\mathbf{k}}/(\omega - E_{-\mathbf{k}})$. 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 $E_{\pm\mathbf{k}} \rightarrow E_{\mathbf{q}\pm\mathbf{k}} \approx E_{\pm\mathbf{k}} \pm \mathbf{q} \cdot \mathbf{v}_{\mathbf{k}}$

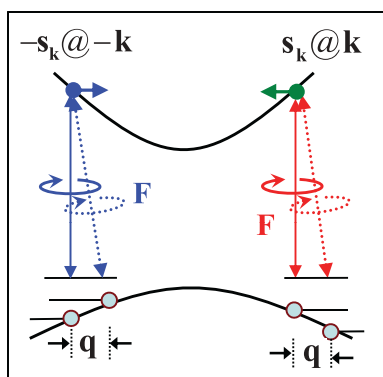


FIG. 2. (Color online) Physical picture for the microscopic mechanism of the *spin current Faraday effect*. The virtual transition energy is the same for $\pm\mathbf{s}_{\mathbf{k}}$ when neglecting light momentum \mathbf{q} .

[see Fig. 2], and $\pm\mathbf{s}_{\mathbf{k}}$ will induce different Faraday rotations due to opposite energy variation. Up to the first order of \mathbf{q} , the polarization is $\mathbf{P}^{(1)} \propto \mathbf{F} \times \mathbf{s}_{\mathbf{k}} \mathbf{v}_{\mathbf{k}} \cdot \mathbf{q}/(\omega - E_{\mathbf{k}})^2$, where $e\mathbf{s}_{\mathbf{k}} \mathbf{v}_{\mathbf{k}}$ is just the spin current tensor contributed by the pair of electrons. This explains the q -dependence of the spin current Faraday effect. More generally, the hole state wave function is also changed when considering the light momentum, which causes extra Berry phase effects [terms proportional to $1/E_F$ in Eq. (37)].

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 twofold degeneracy, and the spin states as well as the selection rules for the interband transitions, like the CB electrons, are independent of the momentum [see Fig. 1(c)].

We first consider a single electron with momentum \mathbf{k} and spin polarization $\mathbf{s}_{\mathbf{k}}$. The spin current contributed by this electron is $\mathbb{J}_{\mathbf{k}} = e\mathbf{s}_{\mathbf{k}} \mathbf{v}_{\mathbf{k}}$ with the velocity $\mathbf{v}_{\mathbf{k}} = \mathbf{k}/m_{\mathbf{e}}$. 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 $s_{\mathbf{k}} = (f_+ - f_-)/2$. The interband transitions $|1/2, \pm 1/2\rangle_{\mathbf{k}} \leftrightarrow |\mp\rangle_{\mathbf{k}}$ couple to a field with circular polarization $\mp(\mathbf{e}_1 \pm i\mathbf{e}_2)/\sqrt{3}$, and the vertical interband transitions $|1/2, \pm 1/2\rangle_{\mathbf{k}} \leftrightarrow |\pm\rangle_{\mathbf{k}}$ 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 interband transitions, the energy shift of this electron due to coupling to an optical field \mathbf{F} is

$$\mathcal{H}_{\text{eff},\mathbf{k}}^{\text{SO}} = -\frac{1}{3} |d_{\text{cv}}|^2 \sum_{\pm} \frac{(1-f_{\pm}) \mathbf{F}^* \cdot (\mathbf{e}_1 \mp i\mathbf{e}_2) (\mathbf{e}_1 \mp i\mathbf{e}_2)^* \cdot \mathbf{F}}{\omega_q - E_{t,-\mathbf{k}+\mathbf{q}} - E_{e\mathbf{k}}} - \frac{1}{3} |d_{\text{cv}}|^2 \sum_{\pm} \frac{(1-f_{\pm}) \mathbf{F}^* \cdot \mathbf{e}_3 \mathbf{e}_3^* \cdot \mathbf{F}}{\omega_q - E_{t,-\mathbf{k}+\mathbf{q}} - E_{e\mathbf{k}}}, \quad (28)$$

where the factor $(1 - f_{\pm})$ 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

$$\mathcal{H}_{\text{eff},\mathbf{k}}^{\text{SO}} = +\frac{2}{3} i |d_{\text{cv}}|^2 \frac{s_{\mathbf{k}} \mathbf{F}^* \cdot (\mathbf{e}_1 \mathbf{e}_2 - \mathbf{e}_2 \mathbf{e}_1) \cdot \mathbf{F}}{\omega_q - E_{t,-\mathbf{k}} - E_{e\mathbf{k}}} + \frac{2}{3} i |d_{\text{cv}}|^2 \frac{s_{\mathbf{k}} \mathbf{F}^* \cdot (\mathbf{e}_1 \mathbf{e}_2 - \mathbf{e}_2 \mathbf{e}_1) \cdot \mathbf{F} \mathbf{q} \cdot \mathbf{k}}{m_t (\omega_q - E_{t,-\mathbf{k}} - E_{e\mathbf{k}})^2}. \quad (29)$$

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

$$\mathcal{H}_{\text{eff}}^{\text{SO}} = -\frac{4}{3} |d_{\text{cv}}|^2 \frac{1}{\Delta_t} I_z \mathbf{z} \cdot \mathbf{S} - \frac{4}{3} |d_{\text{cv}}|^2 \frac{m_e}{em_t \Delta_t^2} q I_z \mathbf{z} \cdot \mathbb{J} \cdot \mathbf{z}, \quad (30)$$

where Δ_t 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 momentum, thus, with the trivial background omitted, the effective Hamiltonian can be derived explicitly as

$$\mathcal{H}_{\text{eff}}^{\text{HL}} = |d_{\text{cv}}|^2 [I_x (\mathbf{x}\mathbf{x} - \mathbf{y}\mathbf{y}) + I_y (\mathbf{x}\mathbf{y} + \mathbf{y}\mathbf{x}) + I_z (i\mathbf{x}\mathbf{y} - i\mathbf{y}\mathbf{x})]:$$

$$\sum_{\mu=\pm, \mathbf{p}} \left[f_{\bar{\mu}, \mathbf{p}} \frac{\mathbf{n}_{\bar{\mu}, \mathbf{p}} \mathbf{n}_{\mu, \mathbf{p}}^*}{E_{e\mathbf{q}+\mathbf{p}} + E_{h-\mathbf{p}} - \omega_q} \right] \quad (31a)$$

$$+ \frac{1}{3} f_{\bar{\mu}, \mathbf{p}} \frac{\mathbf{n}_{\bar{\mu}, \mathbf{p}} \mathbf{n}_{\mu, \mathbf{p}}^* + 2\mathbf{z}_{\mathbf{p}} \mathbf{z}_{\mathbf{p}}^*}{E_{e\mathbf{q}+\mathbf{p}} + E_{l-\mathbf{p}} - \omega_q} \quad (31b)$$

$$- \frac{\sqrt{2}}{3} f_{\mu, \mathbf{p}} \frac{\mathbf{n}_{\bar{\mu}, \mathbf{p}} \mathbf{z}_{\mathbf{p}}^* + \mathbf{z}_{\mathbf{p}} \mathbf{n}_{\mu, \mathbf{p}}^*}{E_{e\mathbf{q}+\mathbf{p}} + E_{l-\mathbf{p}} - \omega_q} \Big], \quad (31c)$$

where $\mu_{\mathbf{p}}$ indicates the spin moment quantized along \mathbf{p} . The terms in the equation above stand for different physical processes as follow. Term (31a) accounts for the HH-CB transitions where an (virtually) absorbed photon will be

emitted with the same circular polarization conserving the electron spin. The LH-CB optical transition can be either circularly polarized or linearly polarized. In Eq. (31b), the absorbed and emitted photons in the virtual LH-CB transitions have the same polarization with the electron spin conserved. In Eq. (31c), the optical polarizations involved in the LH-CB absorption and emission are changed, leading to an angular momentum transfer between the light and the electron, while the total angular momentum is still conserved.

The spin-independent population terms such as $f_{+, \mathbf{p}} + f_{-, \mathbf{p}}$ are related to a change in the background refraction index, but not to the spin polarization and spin current, we will drop the spin-independent population terms and keep the spin polarization terms $\mathbf{s}_{\mathbf{p}}$ only in the effective coupling as

$$\mathcal{H}_{\text{eff}}^{\text{HL}} = |d_{\text{cv}}|^2 [I_x (\mathbf{x}\mathbf{x} - \mathbf{y}\mathbf{y}) + I_y (\mathbf{x}\mathbf{y} + \mathbf{y}\mathbf{x}) + i I_z (\mathbf{x}\mathbf{y} - \mathbf{y}\mathbf{x})]:$$

$$i \sum_{\mathbf{p}} \left[\left(\frac{f_{\mathbf{z}_{\mathbf{p}}, \mathbf{p}+\mathbf{q}} (\mathbf{x}_{\mathbf{p}} \mathbf{y}_{\mathbf{p}} - \mathbf{y}_{\mathbf{p}} \mathbf{x}_{\mathbf{p}})}{E_{e, \mathbf{q}+\mathbf{p}} + E_{h, \mathbf{p}} - \hbar\omega_q} - \frac{1}{3} (E_h \rightarrow E_l) \right) \right.$$

$$\left. + \frac{2}{3} \frac{f_{\mathbf{x}_{\mathbf{p}}, \mathbf{p}+\mathbf{q}} (\mathbf{y}_{\mathbf{p}} \mathbf{z}_{\mathbf{p}} - \mathbf{z}_{\mathbf{p}} \mathbf{y}_{\mathbf{p}}) + f_{\mathbf{y}_{\mathbf{p}}, \mathbf{p}+\mathbf{q}} (\mathbf{z}_{\mathbf{p}} \mathbf{x}_{\mathbf{p}} - \mathbf{x}_{\mathbf{p}} \mathbf{z}_{\mathbf{p}})}{E_{e, \mathbf{p}+\mathbf{q}} + E_{l, \mathbf{p}} - \hbar\omega_q} \right], \quad (32)$$

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

$$\mathcal{H}_{\text{eff}}^{\text{HL}} = -|d_{\text{cv}}|^2 I_z (\mathbf{x}\mathbf{y} - \mathbf{y}\mathbf{x}) : \sum_{\mathbf{p}} \left[\frac{\mathbf{s}_{\mathbf{p}+\mathbf{q}} \cdot \mathbf{z}_{\mathbf{p}} \mathbf{z}_{\mathbf{p}} \cdot \mathcal{E}}{E_{e\mathbf{q}+\mathbf{p}} + E_{h\mathbf{p}} - \hbar\omega_q} - \frac{\mathbf{s}_{\mathbf{p}+\mathbf{q}} \cdot \mathbf{z}_{\mathbf{p}} \mathbf{z}_{\mathbf{p}} \cdot \mathcal{E}}{E_{e\mathbf{q}+\mathbf{p}} + E_{l\mathbf{p}} - \hbar\omega_q} + \frac{2}{3} \frac{\mathcal{E}}{E_{e\mathbf{q}+\mathbf{p}} + E_{l\mathbf{p}} - \hbar\omega_q} \right] \quad (33a)$$

$$= 2 |d_{\text{cv}}|^2 I_z \sum_{\mathbf{p}} \left[\left(\frac{\mathbf{s}_{\mathbf{p}+\mathbf{q}} \cdot \mathbf{z}_{\mathbf{p}} \mathbf{z}_{\mathbf{p}} \cdot \mathbf{z}}{E_{e\mathbf{q}+\mathbf{p}} + E_{h\mathbf{p}} - \hbar\omega_q} - (E_h \rightarrow E_l) \right) + \frac{2}{3} \frac{\mathbf{s}_{\mathbf{p}+\mathbf{q}} \cdot \mathbf{z}}{E_{e\mathbf{q}+\mathbf{p}} + E_{l\mathbf{p}} - \hbar\omega_q} \right]. \quad (33b)$$

By expanding to the first order of \mathbf{q} , we have $E_{e\mathbf{q}+\mathbf{p}} \approx E_{e\mathbf{p}} + \mathbf{q} \cdot \nabla_{\mathbf{p}} E_{e\mathbf{p}}$, and $\mathbf{s}_{\mathbf{p}+\mathbf{q}} \approx \mathbf{s}_{\mathbf{p}} + \mathbf{q} \cdot \nabla_{\mathbf{p}} \mathbf{s}_{\mathbf{p}}$. By using $\nabla_{\mathbf{p}} \mathbf{z}_{\mathbf{p}} = \nabla_{\mathbf{p}} (\mathbf{p}/p) = I^{(2)}/p - \mathbf{p}\mathbf{p}/p^3 = (\mathbf{x}_{\mathbf{p}} \mathbf{x}_{\mathbf{p}} + \mathbf{y}_{\mathbf{p}} \mathbf{y}_{\mathbf{p}})/p$, and $\nabla_{\mathbf{p}} (\mathbf{z}_{\mathbf{p}} \mathbf{z}_{\mathbf{p}}) = (\mathbf{x}_{\mathbf{p}} \mathbf{z}_{\mathbf{p}} \mathbf{x}_{\mathbf{p}} + \mathbf{y}_{\mathbf{p}} \mathbf{z}_{\mathbf{p}} \mathbf{y}_{\mathbf{p}} + \mathbf{x}_{\mathbf{p}} \mathbf{x}_{\mathbf{p}} \mathbf{z}_{\mathbf{p}} + \mathbf{y}_{\mathbf{p}} \mathbf{y}_{\mathbf{p}} \mathbf{z}_{\mathbf{p}})/p$, we obtain the effective Hamiltonian as

$$\mathcal{H}_{\text{eff}}^{\text{HL}} = 2 |d_{\text{cv}}|^2 I_z \sum_{\mathbf{p}} \left[(\mathbf{s}_{\mathbf{p}} \cdot \mathbf{z}_{\mathbf{p}} \mathbf{z}_{\mathbf{p}} \cdot \mathbf{z}) \left(\frac{1}{\Delta_h} - \frac{1}{\Delta_l} \right) + \frac{2}{3} \mathbf{s}_{\mathbf{p}} \cdot \mathbf{z} \frac{1}{\Delta_l} \right] - \frac{2}{e} |d_{\text{cv}}|^2 q I_z \sum_{\mathbf{p}} \text{Tr}[\mathbb{J}_{\mathbf{p}}] \left(\frac{1}{2\Delta_h E_F} - \frac{1}{2\Delta_l E_F} \right)$$

$$+ \frac{2}{e} |d_{\text{cv}}|^2 q I_z \mathbf{z} \mathbf{z} : \sum_{\mathbf{p}} \left[\mathbf{z}_{\mathbf{p}} \mathbf{z}_{\mathbf{p}} \cdot \mathbb{J}_{\mathbf{p}} \left(\frac{m_e}{m_h \Delta_h^2} - \frac{m_e}{m_l \Delta_l^2} + \frac{1}{\Delta_h E_F} - \frac{1}{\Delta_l E_F} \right) + \mathbb{J}_{\mathbf{p}} \left(\frac{2}{3} \frac{m_e}{m_l \Delta_l^2} + \frac{1}{2\Delta_l E_F} - \frac{1}{2\Delta_h E_F} \right) \right], \quad (34)$$

where E_F is the Fermi energy, $\Delta_{h/l}$ 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 ($m_t \rightarrow m_h$, $\Delta_t \rightarrow \Delta_h$). 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 wave vector k_F deviate only slightly from the equilibrium distribution. More specifically, we suppose the spin distribution has the form

$$\mathbf{s}_{\mathbf{p}} = N_0 + N_1 f(p) \cos \theta_{\mathbf{p}}, \quad (35)$$

where $\theta_{\mathbf{p}}$ is the angle between the momentum \mathbf{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]

$$\mathcal{H}_{\text{eff}}^{(0)} = \zeta_0 I_z \mathbf{z} \cdot \mathbf{S}, \quad (36a)$$

$$\mathcal{H}_{\text{eff}}^{(1)} = \zeta_1 q I_z J_Z + \zeta_2 q I_z \mathbf{z} \cdot \mathbb{J} \cdot \mathbf{z}, \quad (36b)$$

with the coupling constants

$$\zeta_0 \equiv \frac{2}{3} |d_{\text{cv}}|^2 \left(\frac{1}{\Delta_h} + \frac{1}{\Delta_l} - \frac{2}{\Delta_t} \right), \quad (37a)$$

$$\zeta_1 \equiv \frac{|d_{\text{cv}}|^2}{e} \left(\frac{2m_e}{5m_h \Delta_h^2} - \frac{2m_e}{5m_l \Delta_l^2} - \frac{3}{5\Delta_h E_F} + \frac{3}{5\Delta_l E_F} \right), \quad (37b)$$

$$\zeta_2 \equiv \frac{|d_{\text{cv}}|^2}{e} \left(\frac{4m_e}{5m_h \Delta_h^2} + \frac{8m_e}{15m_l \Delta_l^2} - \frac{4m_e}{3m_t \Delta_t^2} - \frac{1}{5\Delta_h E_F} + \frac{1}{5\Delta_l E_F} \right). \quad (37c)$$

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 $\mathbf{F}_2 \cdot \mathbf{v}/\mathbf{q} \cdot \mathbf{v}$. 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.^{46–48} If otherwise measured in linear optics, the effect of the chirality relies on the small magnetic moment of the molecules, and in turn on the small wave vector of the probe light, similar to the case of linear optical effects of spin currents.²²

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

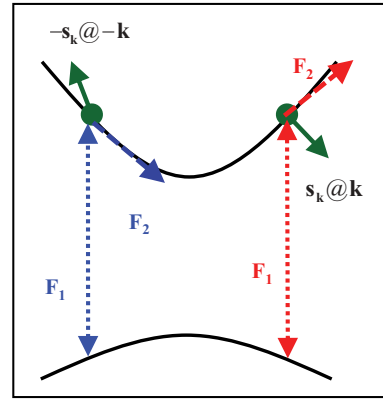


FIG. 3. (Color online) Physical picture for the microscopic mechanism of the second-order nonlinear optical effects of a pure spin current. The second light \mathbf{F}_2 will accelerate the electrons (or holes).

will induce a Faraday rotation $\mathbf{P}^{(1)} \propto \mathbf{F} \times \mathbf{s}_{\mathbf{k}}/(\omega - E_{\mathbf{k}})$. The Faraday rotations due to the pair of spins of $\mathbf{s}_{\mathbf{k}}$ (at momentum \mathbf{k}) and $-\mathbf{s}_{\mathbf{k}}$ (at momentum $-\mathbf{k}$) 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 \mathbf{F}_2 . The spin will experience an intraband acceleration by this optical field and the transition energy will be changed to $E_{\pm\mathbf{k}} \rightarrow E_{\pm\mathbf{k}} \pm \int e\mathbf{v}_{\mathbf{k}} \cdot \mathbf{F}_2 e^{-i\omega_2 t} dt$ [see Fig. 3]. The physical meaning of $e\mathbf{v}_{\mathbf{k}} \cdot \mathbf{F}_2$ is clear that is the power done by the field to the electron. Therefore $\pm\mathbf{s}_{\mathbf{k}}$ will induce different Faraday rotation due to opposite energy modification

$$\mathbf{P}^{(2)} \propto \mathbf{F}_1 \times \mathbf{s}_{\mathbf{k}} e\mathbf{v}_{\mathbf{k}} \cdot \mathbf{F}_2 / [(\omega_1 + \omega_2 - E_{\mathbf{k}})(\omega_1 - E_{\mathbf{k}})\omega_2]. \quad (38)$$

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 reads⁴³

$$\hat{\mathbf{P}}_{\text{intra}}(\mathbf{r}) = ie \sum_{\mathbf{k}, \mathbf{p}} \left[\sum_{\mu, \mu' = \pm} \hat{e}_{\mu'}^{\dagger} \hat{e}_{\mu, \mathbf{k}} \langle \mu' | \mathbf{p} | \mu \rangle_{\mathbf{k}} + \sum_{j', m'; j, m} \hat{V}_{j', m'}^{\dagger} \hat{V}_{j, m; \mathbf{k}} \langle j', m' | \mathbf{p} | j, m \rangle_{\mathbf{k}} \right] \nabla_{\mathbf{k}} e^{i\mathbf{p} \cdot \mathbf{r} - i\mathbf{k} \cdot \mathbf{r}}. \quad (39)$$

With the input optical field consisting of several frequency components $\mathbf{F}(\mathbf{r}, t) = \sum_{j=1,2} \mathbf{F}_j e^{-i\omega_j t} + \text{c.c.}$, the light-matter interaction Hamiltonian is $\hat{H}_1(t) = -\int \hat{\mathbf{P}}(\mathbf{r}) \cdot \mathbf{F}(\mathbf{r}, t) d\mathbf{r}$, where $\hat{\mathbf{P}}(\mathbf{r}) = \hat{\mathbf{P}}(\mathbf{r})_{\text{inter}} + \hat{\mathbf{P}}(\mathbf{r})_{\text{intra}}$. Explicitly, we can write

$$\hat{H}_1(t) = -(\hat{\mathbf{D}} + \hat{\mathbf{D}}^{\dagger} + \hat{\mathbf{d}}) \cdot \left(\sum_{j=1,2} \mathbf{F}_j e^{-i\omega_j t} + \text{c.c.} \right), \quad (40)$$

with

$$\begin{aligned} \hat{\mathbf{D}} \equiv & -d_{\text{cv}}^* \sum_{\mu, \mathbf{k}} (\mathbf{n}_{\bar{\mu}, \mathbf{k}} \hat{h}_{\bar{\mu}, -\mathbf{k}} \hat{e}_{\mu, \mathbf{k}} + (1/\sqrt{3}) \mathbf{n}_{\mu, \mathbf{k}} \hat{l}_{\mu, -\mathbf{k}} \hat{e}_{\mu, \mathbf{k}} \\ & - \sqrt{2/3} \mathbf{z}_{\mathbf{k}} \hat{l}_{\bar{\mu}, -\mathbf{k}} \hat{e}_{\mu, \mathbf{k}} - \mu \sqrt{2/3} \mathbf{n}_{\mu, \mathbf{k}} \hat{l}_{\mu, -\mathbf{k}} \hat{e}_{\mu, \mathbf{k}} \\ & + (\mu/\sqrt{3}) \mathbf{z}_{\mathbf{k}} \hat{l}_{\bar{\mu}, -\mathbf{k}} \hat{e}_{\mu, \mathbf{k}}), \end{aligned} \quad (41a)$$

$$\begin{aligned} \hat{\mathbf{d}} \equiv & ie \sum_{\mathbf{k}, \mathbf{p}} \left(\sum_{\mu=\pm} \hat{e}_{\mu, \mathbf{p}}^\dagger \hat{e}_{\mu, \mathbf{k}} + \sum_{j, m} \hat{V}_{j, m; \mathbf{p}}^\dagger \hat{V}_{j, m; \mathbf{k}} \right) \nabla_{\mathbf{k}} \delta_{\mathbf{p}, \mathbf{k}} \\ & - ie \sum_{\mathbf{k}} \left(\sum_{\mu, \mu'=\pm} \hat{e}_{\mu', \mathbf{k}}^\dagger \hat{e}_{\mu, \mathbf{k}} \langle \mu' |_{\mathbf{k}} \nabla_{\mathbf{k}} | \mu \rangle_{\mathbf{k}} \right. \\ & \left. + \sum_{j, m, m'} \hat{V}_{j, m'; \mathbf{k}}^\dagger \hat{V}_{j, m; \mathbf{k}} \langle j, m' |_{\mathbf{k}} \nabla_{\mathbf{k}} | j, m \rangle_{\mathbf{k}} \right), \end{aligned} \quad (41b)$$

denoting the inter- and intraband polarization operators, respectively. \hat{D} and \hat{D}^\dagger are the positive- and negative-frequency components of the interband polarization operator, respectively. The first part of the intraband 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 intraband polarization. The explicit form of the Berry connection term depends on the choice of the local coordinate $(\mathbf{x}_{\mathbf{p}}, \mathbf{y}_{\mathbf{p}}, \mathbf{z}_{\mathbf{p}})$ 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

$$\mathbf{P}^{(2)}(t) = - \int_{-\infty}^t dt' \int_{-\infty}^{t'} dt'' \text{Tr}[\hat{\mathbf{D}}(t) [\hat{H}_1(t'), [\hat{H}_1(t''), \hat{\rho}_0]]],$$

where $\hat{\mathbf{D}}(t)$, \hat{H}_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 $\hat{\mathbf{D}}(t)$ dominates the optical process; (2) the intraband dipole moment must be considered for the contribution by the spin current; and (3) no holes exist in the initial system, so the interband excitation has to be involved (caused by \hat{D}^\dagger). With all these considerations taken into account, the second-order response of interest is

$$\mathbf{P}^{(2)}(t) = - \int_{-\infty}^t dt' \int_{-\infty}^{t'} dt'' e^{-i\omega_2 t' - i\omega_1 t''} \text{Tr}(\hat{\mathbf{D}}(t) \mathbf{F}_2 \cdot \hat{\mathbf{D}}^\dagger(t') [\mathbf{F}_1 \cdot \hat{\mathbf{d}}(t''), \hat{\rho}_0]) + \{\mathbf{F}_1, \omega_1 \leftrightarrow \mathbf{F}_2, \omega_2\} \quad (42a)$$

$$- \int_{-\infty}^t dt' \int_{-\infty}^{t'} dt'' e^{-i\omega_2 t'' - i\omega_1 t'} \text{Tr}(\hat{\mathbf{D}}(t) [\mathbf{F}_1 \cdot \hat{\mathbf{d}}(t'), \mathbf{F}_2 \cdot \hat{\mathbf{D}}^\dagger(t'') \hat{\rho}_0]) + \{\mathbf{F}_1, \omega_1 \leftrightarrow \mathbf{F}_2, \omega_2\}. \quad (42b)$$

The physical meaning of Eq. (42) is clear: Eq. (42a) corresponds to the driving of the electron population (at t'') followed by interband 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. 23, 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 $\mathbf{P}^{(2)}$ 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 Supplementary Material.⁴⁹

IV. DISCUSSIONS AND NUMERICS

A. Faraday rotation of a spin current and spin polarization

The Faraday rotation angle is expressed as

$$\theta_{\text{F}} = \omega_l l (\chi_{++} - \chi_{--}) / (4nc), \quad (43)$$

where l is the light propagation distance, n is the material refractive index, and c is the light velocity in vacuum [see Appendix C].

Pure spin current. For a spin current configuration as shown in Fig. 4, where a light comes in with a zenith angle β and an

azimuth angle γ , the Faraday rotation angle due to different components of \mathbf{JZ} is

$$\theta_{\text{F}}^{(1)}(J_X) = \delta_{\text{F}}^{(1)} J_X \zeta_2 \sin \beta \cos \gamma, \quad (44a)$$

$$\theta_{\text{F}}^{(1)}(J_Y) = -\delta_{\text{F}}^{(1)} J_Y \zeta_2 \sin^2 \beta \sin \gamma \cos \gamma (n^2 - \sin^2 \beta)^{-1/2}, \quad (44b)$$

$$\theta_{\text{F}}^{(1)}(J_Z) = \delta_{\text{F}}^{(1)} J_Z (\zeta_1 n^2 + \zeta_2 \sin^2 \beta \cos^2 \gamma / n) (n^2 - \sin^2 \beta)^{-1/2}, \quad (44c)$$

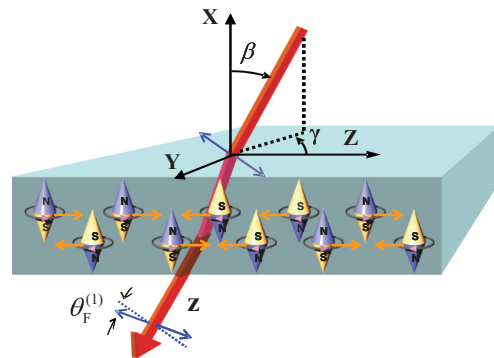


FIG. 4. (Color online) The geometry for measuring a spin current in which the spin current is along Z direction and the red arrow denotes the propagation direction of the light beam.

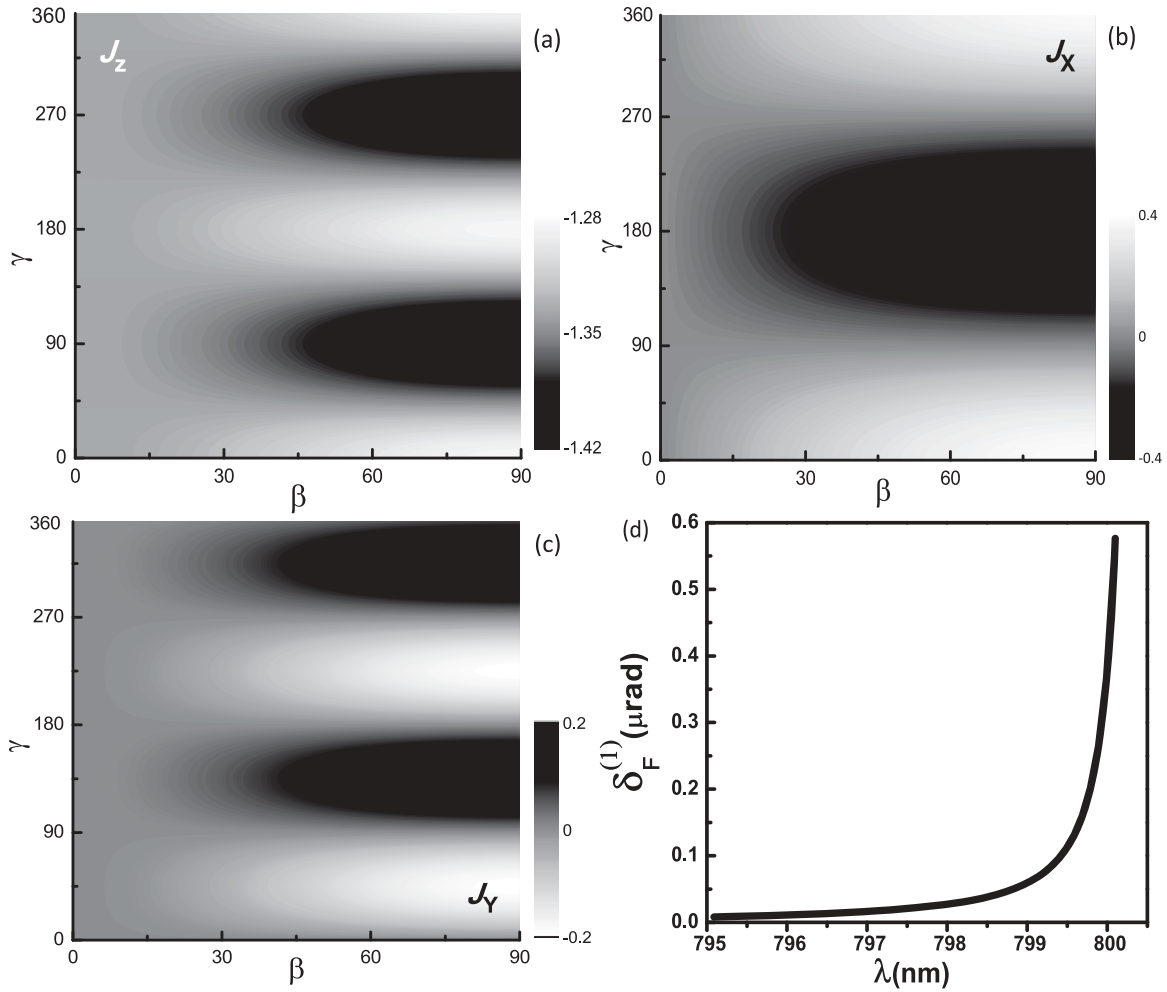


FIG. 5. (a)–(c) The Faraday rotation amplitude of spin current components J_Z , J_Y , and J_X as functions of the incident angles of the light beam. (d) The dependence of $\delta_F^{(1)}$ on the light wavelength λ . Parameters are chosen similar to those in Ref. 8: $E_g = 1519$ meV, $E_{SO} = 341$ meV, the doping concentration is $3 \times 10^{16} \text{ cm}^{-3}$, the effective mass (in units of free electron mass) of the HH, LH, SO, and conduction bands is in turn 0.45, 0.082, 0.15, and 0.067, the dipole $d_{cv} = 6.7 e\text{\AA}$, $n = 3.0$, $L = 2.0 \mu\text{m}$, $E_F = 5.3$ meV, and $J_X = J_Y = J_Z = 20 \text{ nA}/\mu\text{m}^{-2}$.

where $\delta_F^{(1)} = \pi^2 l / 2n\epsilon_0 \lambda^2$. 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 Figs. 5(a)–5(c).

Net spin polarization. The net spin polarization also causes the Faraday rotation. With the incident light of zenith angle β , the Faraday rotation angle equals

$$\theta_F^{(0)}(\mathbf{S}) = (2\pi l / 8\epsilon_0 n \lambda) (\zeta_0 \mathbf{z} \cdot \mathbf{S}). \quad (45)$$

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 β ,

$$\theta_F^{(0)}(\mathbf{S}_\perp) = \pi \zeta_0 S_\perp L / 4\epsilon_0 n \lambda, \quad (46)$$

while for parallel component \mathbf{S}_\parallel ,

$$\theta_F^{(0)}(\mathbf{S}_\parallel) = (\pi \zeta_0 S_\parallel L / 4\epsilon_0 \lambda) \sin \beta \cos \gamma (n^2 - \sin^2 \beta)^{-1/2}. \quad (47)$$

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 $\theta_F^{(0)}(\mathbf{S}_\parallel)$ and $\theta_F^{(1)}(J_X)$ 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 ($\mathcal{P} = -$, odd) and a spin polarization ($\mathcal{P} = +$, 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 $\mathbf{Z} \cdot \mathbf{z} = 0$ and $J_Z = 0$.²² With the experimental configuration shown in Fig. 4, the rotation angle $\theta_F^{(1)}(\beta, \gamma) \propto \delta_F^{(1)} \sin \beta \cos \gamma$. The maximum Faraday rotation angle 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. 5(d). For the specific example shown in Fig. 5(d) with light wavelength around 800 nm, the maximum Faraday rotation angle is 0.38 μrad . 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

$$H_{LK}^A = \frac{1}{2m_0} [(\gamma_1 + 5\gamma_2/2)\nabla^2 - 2\gamma_3(\nabla \cdot \mathbf{K})^2 + 2(\gamma_3 - \gamma_2)(\nabla_x^2 K_x^2 + \text{c.p.})], \quad (48)$$

where the $(\gamma_3 - \gamma_2)$ term describes the anisotropy. The anisotropy is usually small. The eigenfunctions of H_{LK}^A are

$$|\psi_i\rangle = \sum_{j=\pm 3/2, \pm 1/2} \alpha_i^j |3/2, j\rangle, \quad i = \pm 3, \pm 1, \quad (49)$$

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

$$\hat{\mathbf{P}}(\mathbf{r}) = -e \sum_{\mu, \mathbf{k}, \mathbf{p}} e^{-i\mathbf{p}\cdot\mathbf{r} + i\mathbf{k}\cdot\mathbf{r}} \left[\hat{h}_{-, -\mathbf{p}} \hat{e}_{\mu, \mathbf{k}} \langle \psi_3 | \mathbf{r} | \mu \rangle + \hat{h}_{-, -\mathbf{p}} \hat{e}_{\mu, \mathbf{k}} \langle \psi_1 | \mathbf{r} | \mu \rangle + \hat{h}_{-, -\mathbf{p}} \hat{e}_{\mu, \mathbf{k}} \langle \psi_{-1} | \mathbf{r} | \mu \rangle + \hat{h}_{+, -\mathbf{p}} \hat{e}_{\mu, \mathbf{k}} \langle \psi_{-3} | \mathbf{r} | \mu \rangle \right] + \text{H.c.}, \quad (50)$$

where $|\mu\rangle = |\pm\rangle$ denotes the CB electron state with spin $\pm 1/2$, and the operators \hat{h}_{\mp} and \hat{e}_{\mp} annihilate $|\psi_{\pm 3}\rangle$ and $|\psi_{\pm 1}\rangle$, respectively. By using the fact $\mathbf{p}/m_0 = d\mathbf{r}/dt = (\mathbf{r}H_0 - H_0\mathbf{r})/i$, we get

$$\langle \psi_i | -e\mathbf{r} | \mu \rangle = \sum_{j=x, y, z} M_{i, \mathbf{p}} A_{i, \mu}^j \mathbf{j}_p, \quad (51)$$

where $M_{\pm 3/\pm 1, \mathbf{p}} = -ie/m_0(E_{e, \mathbf{p}} - E_{h/l, \mathbf{p}})$. The detailed expression for $A_{i, \mu}^j$ can be found in Appendix C. The effective Hamiltonian then reads

$$\mathcal{H}_{\text{eff}}^A = \text{Tr} \left(\hat{\rho} \sum_{\sigma, \sigma', \mathbf{p}, i, \mu, \mu'}^{\mathbf{j}, \mathbf{j}' = x, y, z} F_{\sigma}^* F_{\sigma'} \mathbf{n}_{\sigma'} \mathbf{n}_{\sigma}^* : |M_{i, \mathbf{p}}|^2 A_{i, \mu}^j A_{i, \mu'}^{j'} \mathbf{j}_p \mathbf{j}_{p'} \frac{1 - f_{\mu\mu', \mathbf{p}+\mathbf{q}}}{E_{e, \mathbf{p}+\mathbf{q}} + E_{i, \mathbf{p}} - \hbar\omega_{\mathbf{q}}} \right). \quad (52)$$

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

$$\begin{aligned} & (|A_{3,+}^1|^2 \mathbf{x}_p \mathbf{x}_p + |A_{3,+}^2|^2 \mathbf{y}_p \mathbf{y}_p + |A_{3,+}^3|^2 \mathbf{z}_p \mathbf{z}_p) (f_{+, \mathbf{p}} + f_{-, \mathbf{p}}) \\ & + 2i [\Im(A_{3,+}^1 A_{3,+}^{2*}) (\mathbf{x}_p \mathbf{y}_p - \mathbf{y}_p \mathbf{x}_p) \\ & + 2i \Im(A_{3,+}^2 A_{3,+}^{3*}) (\mathbf{y}_p \mathbf{z}_p - \mathbf{z}_p \mathbf{y}_p) \\ & + 2i \Im(A_{3,+}^3 A_{3,+}^{1*}) (\mathbf{z}_p \mathbf{x}_p - \mathbf{x}_p \mathbf{z}_p)] f_{z_p}. \end{aligned} \quad (53a)$$

The term (53a) is just a background. The term (53b) 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 E. 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

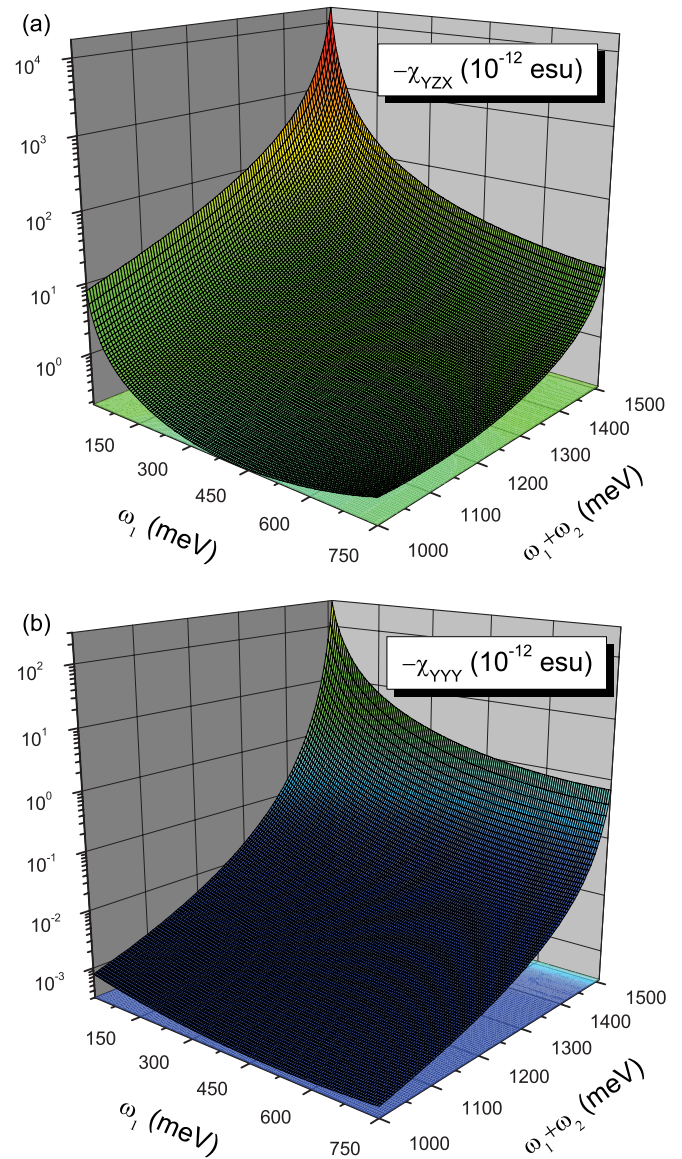


FIG. 6. (Color online) Representative results of the sum frequency susceptibility. (a) $-\chi_{YZX}^{(2)}$ due to a longitudinal spin current and (b) $-\chi_{YYY}^{(2)}$ due to a transverse spin current, as functions of the optical frequencies. Parameters are chosen similar to those in Ref. 8 (same as in Fig. 5). The dielectric constant $\epsilon_r = 10.6$ and the spin current $J_X = J_Z = 20 \text{ nA}/\mu\text{m}^2$.

tensor (not shown) have similar frequency dependence and comparable amplitudes. As a specific example, a transverse spin current $20 \text{ nA}/\mu\text{m}^2$ has a susceptibility $-\chi_{YY}^{(2)} \approx 4.8 \times 10^{-12} \text{ esu}$ (or $0.2 \times 10^{-14} \text{ m/V}$ 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 current can be straightforwardly extended to other second-order optical spectroscopy such as difference frequency and three-wave mixing.⁴⁰

V. CONCLUSIONS

In summary, with the systematic symmetry analysis 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.⁵⁰⁻⁵⁵ A wealth of physics connecting spins and photons and technologies synthesizing spintronics and photonics may be explored.

ACKNOWLEDGMENTS

This work was supported by Hong Kong RGC/GRF CUHK 401011, the NSFC Grants No. 11074143, No. 10774086, and No. 10574076 and the Basic Research Program of China Grants No. 2006CB921500 and No. 2011CB921901.

APPENDIX A: COORDINATE BASIS

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

$$\hat{\theta}_p = \mathbf{x}_p = \mathbf{X} \cos \theta_p \cos \phi_p + \mathbf{Y} \cos \theta_p \sin \phi_p - \mathbf{Z} \sin \theta_p, \quad (\text{A1a})$$

$$\hat{\phi}_p = \mathbf{y}_p = -\mathbf{X} \sin \phi_p + \mathbf{Y} \cos \phi_p, \quad (\text{A1b})$$

$$\hat{\mathbf{p}} = \mathbf{z}_p = \mathbf{X} \sin \theta_p \cos \phi_p + \mathbf{Y} \sin \theta_p \sin \phi_p + \mathbf{Z} \cos \theta_p. \quad (\text{A1c})$$

The angle average of the tensor

$$\overline{\mathbf{z}_p \mathbf{z}_p} \equiv \frac{1}{4\pi} \int \mathbf{z}_p \mathbf{z}_p d\Omega = \frac{1}{3} I^{(2)}. \quad (\text{A2})$$

And the angle average

$$\begin{aligned} & \overline{\mathbf{Z} \cdot \mathbf{z}_p \mathbf{z}_p \mathbf{z}_p \mathbf{z}_p} \\ & \equiv \frac{1}{4\pi} \int \mathbf{Z} \cdot \mathbf{z}_p \mathbf{z}_p \mathbf{z}_p \mathbf{z}_p d\Omega \\ & = \overline{\cos^2 \theta_p \sin^2 \theta_p \cos^2 \phi_p (\mathbf{X}\mathbf{X}\mathbf{Z} + \mathbf{X}\mathbf{Z}\mathbf{X} + \mathbf{Z}\mathbf{X}\mathbf{X})} \\ & \quad + \overline{\cos^2 \theta_p \sin^2 \theta_p \sin^2 \phi_p (\mathbf{Y}\mathbf{Y}\mathbf{Z} + \mathbf{Y}\mathbf{Z}\mathbf{Y} + \mathbf{Z}\mathbf{Y}\mathbf{Y})} \\ & \quad + \overline{\cos^4 \theta_p \mathbf{Z}\mathbf{Z}\mathbf{Z}} \\ & = \frac{1}{15} (I^{(2)} \mathbf{Z} + \mathbf{X}\mathbf{X}\mathbf{Z} + \mathbf{Y}\mathbf{Y}\mathbf{Z} + \mathbf{Z}\mathbf{Z}\mathbf{Z} + \mathbf{Z}I^{(2)}). \quad (\text{A3}) \end{aligned}$$

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

$$\mathbf{S} = \sum_{\mathbf{p}} \mathbf{s}_p = \sum_{\mathbf{p}} \mathbf{N}_0, \quad (\text{A4a})$$

$$\begin{aligned} \mathbb{J} & = \sum_{\mathbf{p}} \mathbb{J}_p = \frac{e}{m_e} \sum_{\mathbf{p}} \mathbf{N}_1 f(p) p \mathbf{Z} \cdot \mathbf{z}_p \mathbf{z}_p \\ & = \frac{e}{m_e} \sum_{\mathbf{p}} \mathbf{N}_1 f(p) p \mathbf{Z} \cdot \overline{\mathbf{z}_p \mathbf{z}_p} \\ & = \frac{\mathbf{N}_1 \mathbf{Z}}{3} \frac{e}{m_e} \sum_{\mathbf{p}} f(p) p = \mathbf{J}\mathbf{Z}. \quad (\text{A4b}) \end{aligned}$$

Also, we have

$$\begin{aligned} \sum_{\mathbf{p}} \mathbf{z}_p \mathbf{z}_p \cdot \mathbb{J}_p & = \frac{e}{m_e} \sum_{\mathbf{p}} \mathbf{z}_p \mathbf{z}_p \cdot (\mathbf{N}_1 f(p) p \mathbf{Z} \cdot \mathbf{z}_p \mathbf{z}_p) \\ & = \frac{e}{m_e} \sum_{\mathbf{p}} f(p) p \overline{\mathbf{z}_p \mathbf{z}_p \mathbf{z}_p \mathbf{z}_p} \cdot \mathbf{N}_1 \\ & = \frac{1}{3} \frac{e}{m_e} \sum_{\mathbf{p}} f(p) p \\ & \quad \times \frac{I^{(2)} \mathbf{Z} + \mathbf{X}\mathbf{Z}\mathbf{X} + \mathbf{Y}\mathbf{Z}\mathbf{Y} + \mathbf{Z}\mathbf{Z}\mathbf{Z} + \mathbf{Z}I^{(2)}}{3} \cdot \mathbf{N}_1 \\ & = \frac{1}{5} (J_Z I^{(2)} + \mathbb{J} + \mathbb{J}^T). \quad (\text{A5}) \end{aligned}$$

APPENDIX B: BERRY CONNECTION

The band edge states of CB are

$$|+/-\rangle_{\mathbf{p}} = |S\rangle \otimes |\uparrow/\downarrow\rangle_{\mathbf{p}}, \quad (\text{B1})$$

with $|S\rangle$ being a periodic s -wave orbital wave function, which is isotropic in a unit cell, and $|\uparrow/\downarrow\rangle_{\mathbf{p}}$ denoting the spin eigenstate parallel/antiparallel to the momentum.

Similarly, the band edge states of the valence bands are

$$\left| \frac{3}{2}, +\frac{3}{2} \right\rangle_{\mathbf{p}} = -\frac{|X\rangle_{\mathbf{p}} + i|Y\rangle_{\mathbf{p}}}{\sqrt{2}} \otimes |\uparrow\rangle_{\mathbf{p}}, \quad (\text{B2a})$$

$$\left| \frac{3}{2}, +\frac{1}{2} \right\rangle_{\mathbf{p}} = \sqrt{\frac{2}{3}} |Z\rangle_{\mathbf{p}} \otimes |\uparrow\rangle_{\mathbf{p}} - \frac{|X\rangle_{\mathbf{p}} + i|Y\rangle_{\mathbf{p}}}{\sqrt{6}} \otimes |\downarrow\rangle_{\mathbf{p}}, \quad (\text{B2b})$$

$$\left| \frac{3}{2}, -\frac{1}{2} \right\rangle_{\mathbf{p}} = \sqrt{\frac{2}{3}} |Z\rangle_{\mathbf{p}} \otimes |\downarrow\rangle_{\mathbf{p}} + \frac{|X\rangle_{\mathbf{p}} - i|Y\rangle_{\mathbf{p}}}{\sqrt{6}} \otimes |\uparrow\rangle_{\mathbf{p}}, \quad (\text{B2c})$$

$$\left| \frac{3}{2}, -\frac{3}{2} \right\rangle_{\mathbf{p}} = +\frac{|X\rangle_{\mathbf{p}} - i|Y\rangle_{\mathbf{p}}}{\sqrt{2}} \otimes |\downarrow\rangle_{\mathbf{p}}, \quad (\text{B2d})$$

$$\left| \frac{1}{2}, +\frac{1}{2} \right\rangle_{\mathbf{p}} = -\frac{1}{\sqrt{3}} |Z\rangle_{\mathbf{p}} \otimes |\uparrow\rangle_{\mathbf{p}} - \frac{|X\rangle_{\mathbf{p}} + i|Y\rangle_{\mathbf{p}}}{\sqrt{3}} \otimes |\downarrow\rangle_{\mathbf{p}}, \quad (\text{B2e})$$

$$\left| \frac{1}{2}, -\frac{1}{2} \right\rangle_{\mathbf{p}} = +\frac{1}{\sqrt{3}} |Z\rangle_{\mathbf{p}} \otimes |\downarrow\rangle_{\mathbf{p}} - \frac{|X\rangle_{\mathbf{p}} - i|Y\rangle_{\mathbf{p}}}{\sqrt{3}} \otimes |\uparrow\rangle_{\mathbf{p}}. \quad (\text{B2f})$$

where $|X\rangle_{\mathbf{p}}, |Y\rangle_{\mathbf{p}}, |Z\rangle_{\mathbf{p}}$ are the p -type orbital parts of the Bloch amplitudes with wave vector \mathbf{p} , which have the same rotation and inversion transformation properties as the coordinate system $\mathbf{x}_{\mathbf{p}}, \mathbf{y}_{\mathbf{p}}, \mathbf{z}_{\mathbf{p}}$, defined with respect to the momentum direction (i.e., $\mathbf{z}_{\mathbf{p}} = \mathbf{p}/p$). The mixing of the orbital wave functions and the electron spin states in the total angular momentum eigenstates includes the spin-orbit coupling automatically. This spin-orbit coupling is an intrinsic relativistic effect and does not rely on whether or not the material has inversion symmetry.

With the convention chosen in Eq. (A1), the transformation of the Bloch and spin states are as follows:

$$|X\rangle_{\mathbf{p}} = |X\rangle \cos \theta_{\mathbf{p}} \cos \phi_{\mathbf{p}} + |Y\rangle \cos \theta_{\mathbf{p}} \sin \phi_{\mathbf{p}} - |Z\rangle \sin \theta_{\mathbf{p}}, \quad (\text{B3a})$$

$$|Y\rangle_{\mathbf{p}} = -|X\rangle \sin \phi_{\mathbf{p}} + |Y\rangle \cos \phi_{\mathbf{p}}, \quad (\text{B3b})$$

$$|Z\rangle_{\mathbf{p}} = |X\rangle \sin \theta_{\mathbf{p}} \cos \phi_{\mathbf{p}} + |Y\rangle \sin \theta_{\mathbf{p}} \sin \phi_{\mathbf{p}} + |Z\rangle \cos \theta_{\mathbf{p}}, \quad (\text{B3c})$$

$$|\uparrow\rangle_{\mathbf{p}} = +\cos \frac{\theta_{\mathbf{p}}}{2} e^{-i\phi_{\mathbf{p}}/2} |\uparrow\rangle + \sin \frac{\theta_{\mathbf{p}}}{2} e^{+i\phi_{\mathbf{p}}/2} |\downarrow\rangle, \quad (\text{B3d})$$

$$|\downarrow\rangle_{\mathbf{p}} = -\sin \frac{\theta_{\mathbf{p}}}{2} e^{-i\phi_{\mathbf{p}}/2} |\uparrow\rangle + \cos \frac{\theta_{\mathbf{p}}}{2} e^{+i\phi_{\mathbf{p}}/2} |\downarrow\rangle. \quad (\text{B3e})$$

$|X\rangle, |Y\rangle, |Z\rangle$ are the orbital Bloch functions which transform as X, Y, Z , and $|\uparrow\rangle, |\downarrow\rangle$ are the spin Bloch function as the eigenstates of $\sigma \cdot \mathbf{Z}$ with eigenvalue ± 1 . With this convention, the Berry curvature term has a very simple form as

$$-i\langle j, m' |_{\mathbf{p}} \nabla_{\mathbf{p}} |j, m\rangle_{\mathbf{p}} = i \frac{\mathbf{n}_{\pm, \mathbf{p}}}{\sqrt{2p}} \delta_{m' \pm 1, m} \sqrt{(j \pm m)(j \mp m + 1)} - \delta_{m, m'} m \frac{\cos \theta_{\mathbf{p}}}{\sin \theta_{\mathbf{p}}} \frac{\mathbf{y}_{\mathbf{p}}}{p}. \quad (\text{B4})$$

APPENDIX C: FARADAY ROTATION ANGLE

For a light with frequency ω_q , the polarization density is

$$\mathbf{P} = \epsilon_0 \sum_{\sigma, \sigma'} \mathbf{n}_{\sigma} \chi_{\sigma, \sigma'} F_{\sigma'}. \quad (\text{C1})$$

Then the energy density in the material is

$$\rho_E = \frac{1}{2} \langle \mathbf{D}(\mathbf{r}, t) \cdot \mathbf{F}(\mathbf{r}, t) \rangle + \frac{1}{2} \langle \mathbf{B}(\mathbf{r}, t) \cdot \mathbf{H}(\mathbf{r}, t) \rangle = (\epsilon_0 \epsilon_r \mathbf{F} + \mathbf{P}) \cdot \mathbf{F}^* + \text{c.c.}, \quad (\text{C2})$$

where ϵ_r is the background dielectric constant. Thus the linear optical susceptibility is related to the effective Hamiltonian through

$$\mathcal{H}_{\text{eff}} = \epsilon_0 \sum_{\sigma, \sigma'} \chi_{\sigma, \sigma'} F_{\sigma}^* F_{\sigma'} + \epsilon_0 \sum_{\sigma, \sigma'} \chi_{\sigma, \sigma'}^* F_{\sigma} F_{\sigma'}^*. \quad (\text{C3})$$

Thus we have

$$\chi_{\sigma, \sigma'} + \chi_{\sigma', \sigma}^* = \frac{1}{\epsilon_0} \frac{\partial^2 \mathcal{H}_{\text{eff}}}{\partial F_{\sigma}^* \partial F_{\sigma'}}. \quad (\text{C4})$$

The index change due to two circular polarization is, respectively,

$$\delta n_{\pm} = \sqrt{\epsilon_r + \chi_{\pm\pm}} - \sqrt{\epsilon_r} \approx \pm \frac{1}{2} \chi_{\pm\pm} / \sqrt{\epsilon_r} = \pm \frac{1}{2} n^{-1} \chi_{\pm\pm}, \quad (\text{C5})$$

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

$$\delta \phi_{\pm} = \omega_q c^{-1} l \delta n_{\pm} = 2\pi \lambda^{-1} l \delta n_{\pm}, \quad (\text{C6})$$

where λ is the light wavelength in vacuum. For a light with linear polarization

$$\mathbf{x} = (-\mathbf{n}_+ + \mathbf{n}_-) / \sqrt{2}, \quad (\text{C7})$$

after propagation of the length l , the polarization becomes

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

So the Faraday rotation angle is

$$\theta_F = \delta\phi_+ = \frac{2\pi l}{2n\lambda} \chi_{++}. \quad (\text{C9})$$

For a light with incident zenith and azimuth angles β and γ , the angles inside the sample β' and γ' are determined by

$$n \sin \beta' = \sin \beta, \quad (\text{C10a})$$

$$\gamma' = \gamma, \quad (\text{C10b})$$

the propagation length through a sample of thickness L is

$$l = L / \cos \beta'. \quad (\text{C11})$$

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

$$\chi_{++}^{(1)} = -\chi_{--}^{(1)} = \frac{1}{4\epsilon_0} (\zeta_1 q J_Z + \zeta_2 q \mathbf{z} \cdot \mathbb{J} \cdot \mathbf{z}), \quad (\text{C12})$$

Thus the Faraday rotation for a spin current polarized normal to the surface (as in Awschalom's experiment⁸) is

$$\begin{aligned} \theta_F^{(1)} = \delta\phi_+ &= \frac{2\pi q L}{8\epsilon_0 n \lambda \cos \beta'} \zeta_2 J \cos \beta' \sin \beta' \cos \gamma \\ &= \frac{\pi^2 \zeta_2 J L}{2n\epsilon_0 \lambda^2} \sin \beta \cos \gamma, \end{aligned} \quad (\text{C13})$$

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

APPENDIX D: ANISOTROPIC VALENCE BAND EFFECT

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

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

where

$$E_3 = E_{-3} = \frac{1}{2m_0} [(\gamma_1 + \gamma_2)k^2 - 3\gamma_2 k_z^2], \quad (\text{D2a})$$

$$E_1 = E_{-1} = \frac{1}{2m} [(\gamma_1 - \gamma_2)k^2 + 3\gamma_2 k_z^2], \quad (\text{D2b})$$

$$P = -\frac{\sqrt{3}\gamma_3}{m_0} k_z (k_x - ik_y), \quad (\text{D2c})$$

$$Q = \frac{1}{2m_0} [-\sqrt{3}\gamma_2 (k_x^2 - k_y^2) + i2\sqrt{3}\gamma_3 k_x k_y]. \quad (\text{D2d})$$

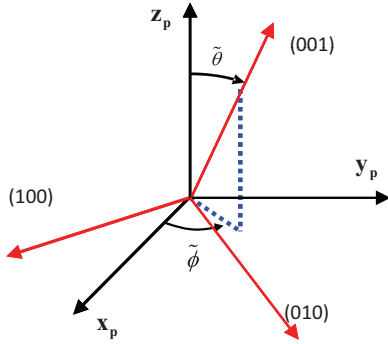


FIG. 7. (Color online) The geometry of the coordinates.

The eigenstates can be in general written as

$$|\psi_i\rangle = \sum_{j=\pm\frac{3}{2}, \pm\frac{1}{2}} \alpha_i^j \left| \frac{3}{2}, j \right\rangle, \quad i = \pm 3, \pm 1. \quad (\text{D3})$$

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

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

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

$$U^\dagger \alpha = \alpha^*. \quad (\text{D5})$$

Without loss of generality, we take the coordinate relation between the electron and the crystal as [see Fig. 7]

$$\begin{aligned} (100) &= \sin \tilde{\phi} \tilde{x}_p - \cos \tilde{\phi} \tilde{y}_p, \\ (010) &= \cos \tilde{\theta} \cos \tilde{\phi} \tilde{x}_p + \cos \tilde{\theta} \sin \tilde{\phi} \tilde{y}_p - \sin \tilde{\theta} \tilde{z}_p, \\ (001) &= \sin \tilde{\theta} \cos \tilde{\phi} \tilde{x}_p + \sin \tilde{\theta} \sin \tilde{\phi} \tilde{y}_p + \cos \tilde{\theta} \tilde{z}_p, \end{aligned} \quad (\text{D6})$$

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

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

$$A_{i,\pm}^x = \left[\mp \frac{1}{\sqrt{2}} (\alpha_i^{\pm\frac{3}{2}})^* \pm \frac{1}{\sqrt{6}} (\alpha_i^{\mp\frac{1}{2}})^* \right] \sin \tilde{\phi} + i \left[\frac{1}{\sqrt{2}} (\alpha_i^{\pm\frac{3}{2}})^* + \frac{1}{\sqrt{6}} (\alpha_i^{\mp\frac{1}{2}})^* \right] \cos \tilde{\theta} \cos \tilde{\phi} + \sqrt{\frac{2}{3}} (\alpha_i^{\pm\frac{1}{2}})^* \sin \tilde{\theta} \cos \tilde{\phi}, \quad (\text{D7a})$$

$$A_{i,\pm}^y = \left[\pm \frac{1}{\sqrt{2}} (\alpha_i^{\pm\frac{3}{2}})^* \mp \frac{1}{\sqrt{6}} (\alpha_i^{\mp\frac{1}{2}})^* \right] \cos \tilde{\phi} + i \left[\frac{1}{\sqrt{2}} (\alpha_i^{\pm\frac{3}{2}})^* + \frac{1}{\sqrt{6}} (\alpha_i^{\mp\frac{1}{2}})^* \right] \cos \tilde{\theta} \sin \tilde{\phi} + \sqrt{\frac{2}{3}} (\alpha_i^{\pm\frac{1}{2}})^* \sin \tilde{\theta} \cos \tilde{\phi}, \quad (\text{D7b})$$

$$A_{i,\pm}^z = -i \left[\frac{1}{\sqrt{2}} (\alpha_i^{\pm\frac{3}{2}})^* + \frac{1}{\sqrt{6}} (\alpha_i^{\mp\frac{1}{2}})^* \right] \sin \tilde{\theta} + \sqrt{\frac{2}{3}} (\alpha_i^{\pm\frac{1}{2}})^* \cos \tilde{\theta}, \quad (\text{D7c})$$

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

$$A_{i,\nu}^j = -\nu A_{-i,-\nu}^{j*}. \quad (\text{D8})$$

APPENDIX E: SECOND-ORDER NONLINEAR SUSCEPTIBILITY

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

$$\begin{aligned} \chi^{(2)}(\omega_1, \omega_2; \omega_1 + \omega_2) &= J_X [\mathbf{XXY}(-2\xi_3 - 2\xi_3' + \xi_4' + \xi_5') + \mathbf{ZZY}(-4\xi_3 - \xi_1' + \xi_3' - \xi_5') + \mathbf{YXX}(-\xi_4 - \xi_5 - \xi_4' - \xi_5') \\ &\quad + \mathbf{XYX}(-2\xi_3 + \xi_4 + \xi_5 - 2\xi_3') + \mathbf{ZYZ}(-\xi_1 + \xi_3 - \xi_5 - 4\xi_3') \\ &\quad + \mathbf{YZZ}(\xi_1 - \xi_3 + \xi_5 + \xi_1' - \xi_3' + \xi_5') + \mathbf{YYY}(-4\xi_3 - 4\xi_3')] \end{aligned} \quad (\text{E1a})$$

$$\begin{aligned} &+ J_Z [(\mathbf{XYZ} - \mathbf{YXZ})(\xi_1 + \xi_2 + 2\xi_3 + \xi_4 + 3\xi_5 - \xi_2' - 3\xi_3' - \xi_5') \\ &\quad + (\mathbf{YZX} - \mathbf{XZY})(\xi_2 + 3\xi_3 + \xi_5 - \xi_1' - \xi_2' - 2\xi_3' - \xi_4' - 3\xi_5') \\ &\quad + (\mathbf{ZXY} - \mathbf{ZYX})(\xi_2 + 5\xi_3 + \xi_5 - \xi_2' - 5\xi_3 - \xi_5')], \end{aligned} \quad (\text{E1b})$$

where ξ_k' is derived from ξ_k by exchanging ω_1 and ω_2 , and

$$\xi_1 = \left(\frac{\epsilon_r + 2}{3} \right)^3 |d_{cv}|^2 \frac{2}{3} \left[\frac{1 + m_e/m_l}{(\Delta^l)^2 \omega_1} + \frac{1 + m_e/m_l}{(\Delta^l)^2 \Delta_2^l} - \frac{1 + m_e/m_t}{(\Delta^l)^2 \omega_1} - \frac{1 + m_e/m_t}{(\Delta^l)^2 \Delta_2^l} \right], \quad (\text{E2a})$$

$$\xi_2 = \left(\frac{\epsilon_r + 2}{3} \right)^3 |d_{cv}|^2 \left(\frac{\Delta^l - \Delta^h}{2E_F \Delta^h \Delta^l \omega_1} + \frac{\Delta^l - \Delta^h}{2E_F \Delta^h \Delta^l \Delta_2^l} \right), \quad (\text{E2b})$$

$$\xi_3 = \left(\frac{\epsilon_r + 2}{3}\right)^3 |d_{cv}|^2 \frac{1}{5} \frac{(\Delta^l - \Delta^h)(\Delta_2^l - \Delta_2^h)}{4E_F \Delta^h \Delta_2^h \Delta^l \Delta_2^l}, \quad (\text{E2c})$$

$$\xi_4 = \left(\frac{\epsilon_r + 2}{3}\right)^3 |d_{cv}|^2 \left[\frac{\Delta^l - \Delta^h}{2E_F \Delta^h \Delta^l \omega_1} + \frac{(\Delta^l - \Delta^h)(\Delta_2^l + \Delta_2^h)}{4E_F \Delta^h \Delta_2^h \Delta^l \Delta_2^l} \right], \quad (\text{E2d})$$

$$\xi_5 = \left(\frac{\epsilon_r + 2}{3}\right)^3 \frac{|d_{cv}|^2}{5} \left[\frac{1 + m_e/m_h}{(\Delta^h)^2 \omega_1} + \frac{1 + m_e/m_h}{(\Delta^h)^2 \Delta_2^h} - \frac{1 + m_e/m_l}{(\Delta^l)^2 \omega_1} - \frac{1 + m_e/m_l}{(\Delta^l)^2 \Delta_2^l} - \frac{\Delta^l - \Delta^h}{E_F \Delta^h \Delta^l \omega_1} - \frac{\Delta^l - \Delta^h}{2E_F \Delta^h \Delta^l \Delta_2^l} - \frac{(\Delta^l - \Delta^h)(\Delta_2^l + \Delta_2^h)}{4E_F \Delta^h \Delta_2^h \Delta^l \Delta_2^l} \right], \quad (\text{E2e})$$

where the factor containing the material dielectric constant ϵ_r takes into account the difference between the macroscopic external field and the microscopic local field.⁵⁶

*Current address: Department of Physics, Stanford University, Stanford, California 94305-4045, USA.

[†]bfz@mail.tsinghua.edu.cn

[‡]rbliu@phy.cuhk.edu.hk

¹S. A. Wolf, D. D. Awschalom, R. A. Buhrman, J. M. Daughton, S. von Molnár, M. L. Roukes, A. Y. Chtchelkanova, and D. M. Treger, *Science* **294**, 1488 (2001).

²I. Žutić, J. Fabian, and S. Das Sarma, *Rev. Mod. Phys.* **76**, 323 (2004).

³J. M. Kikkawa and D. D. Awschalom, *Nature (London)* **397**, 139 (1999).

⁴J. Stephens, J. Berezovsky, J. P. McGuire, L. J. Sham, A. C. Gossard, and D. D. Awschalom, *Phys. Rev. Lett.* **93**, 097602 (2004).

⁵S. A. Crooker, M. Furis, X. Lou, C. Adelman, D. L. Smith, C. J. Palmstrøm, and P. A. Crowell, *Science* **309**, 2191 (2005).

⁶X. H. Lou, C. Adelman, S. A. Crooker, E. S. Garlid, J. Zhang, K. S. M. Reddy, S. D. Flexner, C. J. Palmstrøm, and P. A. Crowell, *Nat. Phys.* **3**, 197 (2007).

⁷I. Appelbaum, B. Q. Huang, and D. J. Monsma, *Nature (London)* **447**, 295 (2007).

⁸Y. K. Kato, R. C. Myers, A. C. Gossard, and D. D. Awschalom, *Science* **306**, 1910 (2004).

⁹N. P. Stern, D. W. Steuerman, S. Mack, A. C. Gossard, and D. D. Awschalom, *Nat. Phys.* **4**, 843 (2008).

¹⁰J. Wunderlich, B. Kaestner, J. Sinova, and T. Jungwirth, *Phys. Rev. Lett.* **94**, 047204 (2005).

¹¹M. J. Stevens, A. L. Smirl, R. D. R. Bhat, A. Najmaie, J. E. Sipe, and H. M. van Driel, *Phys. Rev. Lett.* **90**, 136603 (2003).

¹²H. Zhao, E. J. Loren, H. M. van Driel, and A. L. Smirl, *Phys. Rev. Lett.* **96**, 246601 (2006).

¹³M. I. Dyakonov and V. I. Perel, *Phys. Lett. A* **35**, 459 (1971).

¹⁴J. E. Hirsch, *Phys. Rev. Lett.* **83**, 1834 (1999).

¹⁵S. Murakami, N. Nagaosa, and S. C. Zhang, *Science* **301**, 1348 (2003).

¹⁶J. Sinova, D. Culcer, Q. Niu, N. A. Sinitsyn, T. Jungwirth, and A. H. MacDonald, *Phys. Rev. Lett.* **92**, 126603 (2004).

¹⁷S. O. Valenzuela and M. Tinkham, *Nature (London)* **442**, 176 (2006).

¹⁸S. D. Ganichev *et al.*, *Phys. Rev. B* **75**, 155317 (2007).

¹⁹X. D. Cui, S. Q. Shen, J. Li, Y. Ji, W. Ge, and F. C. Zhang, *Appl. Phys. Lett.* **90**, 241125 (2007).

²⁰C. Brüne, A. Roth, E. G. Novik, M. König, H. Buhmann, E. M. Hankiewicz, W. Hanke, J. Sinova, and L. W. Molenkamp, *Nat. Phys.* **6**, 448 (2010).

²¹C. Brüne, A. Roth, H. Buhmann, E. M. Hankiewicz, L. W. Molenkamp, J. Maciejko, X.-L. Qi, and S.-C. Zhang, *Nat. Phys.* **8**, 486 (2012).

²²J. Wang, B. F. Zhu, and R. B. Liu, *Phys. Rev. Lett.* **100**, 086603 (2008), see also Erratum, *ibid.* **101**, 069902 (2008).

²³J. Wang, B. F. Zhu, and R. B. Liu, *Phys. Rev. Lett.* **104**, 256601 (2010).

²⁴S. Coleman, J. Wess, and B. Zumino, *Phys. Rev.* **177**, 2239 (1969).

²⁵C. G. Callan, S. Coleman, J. Wess, and B. Zumino, *Phys. Rev.* **177**, 2247 (1969).

²⁶Strictly speaking, a light beam does not conserve the time-reversal symmetry. But its angular momentum flux apart from the energy flux, being a pure spin current of interest here, does.

²⁷R. C. Jones, *J. Opt. Soc. Am.* **31**, 488 (1941).

²⁸*Faraday's Diary*, edited by Thomas Martin (London: George Bell and Sons, Ltd, Nov. 12, 1839 - June 26, 1847), Vol. IV.

²⁹V. Vlaminck and M. Bailleul, *Science* **322**, 410 (2008).

³⁰Y. A. Bychkov and E. I. Rashba, *Pis'ma Zh. Eksp. Teor. Fiz.* **39**, 66 (1984) [*Sov. Phys. JETP Lett.* **39**, 78 (1984)].

³¹G. Dresselhaus, *Phys. Rev.* **100**, 580 (1955).

³²S. D. Ganichev *et al.*, *Phys. Rev. Lett.* **92**, 256601 (2004).

³³L. K. Weraka and H. Zhao, *Nat. Phys.* **6**, 875 (2010).

³⁴R. D. R. Bhat and J. E. Sipe, *Phys. Rev. Lett.* **85**, 5432 (2000).

³⁵R. D. R. Bhat, F. Nastos, A. Najmaie, and J. E. Sipe, *Phys. Rev. Lett.* **94**, 096603 (2005).

³⁶G. E. Pikus, V. A. Marushchak, and A. N. Titkov, *Sov. Phys. Semicond.* **22**, 115 (1988).

³⁷Q. F. Sun and X. C. Xie, *Phys. Rev. B* **72**, 245305 (2005).

³⁸J. Shi, P. Zhang, D. Xiao, and Q. Niu, *Phys. Rev. Lett.* **96**, 076604 (2006).

³⁹A. K. Zvezdin and V. A. Kotov, *Modern Magneto-optics and Magneto-optical Materials* (Taylor and Francis Group, New York, 1997).

⁴⁰Y. R. Shen, *The Principles of Nonlinear Optics* (Wiley-Interscience, New York, 1984).

⁴¹J. B. Khurgin, *Appl. Phys. Lett.* **67**, 1113 (1995).

- ⁴²B. A. Ruzicka, L. K. Werake, G. Xu, J. B. Khurgin, E. Y. Sherman, J. Z. Wu, and H. Zhao, *Phys. Rev. Lett.* **108**, 077403 (2012).
- ⁴³J. E. Sipe and A. I. Shkrebtii, *Phys. Rev. B* **61**, 5337 (2000).
- ⁴⁴P. Y. Yu and M. Cardona, *Fundamentals of Semiconductors: Physics and Material Properties*, 3rd ed. (Springer-Verlag, Berlin, 2005).
- ⁴⁵W. Paul and T. S. Moss, *Handbook on Semiconductors: Band Theory and Transport Properties*, Vol. 1 (North-Holland, Amsterdam, 1982).
- ⁴⁶M. A. Belkin, T. A. Kulakov, K.-H. Ernst, L. Yan, and Y. R. Shen, *Phys. Rev. Lett.* **85**, 4474 (2000).
- ⁴⁷N. Ji, V. Ostroverkhov, M. Belkin, Y.-J. Shiu, and Y.-R. Shen, *J. Am. Chem. Soc.* **128**, 8845 (2006).
- ⁴⁸J. Wang, X. Chen, M. L. Clarke, and Z. Chen, *Proc. Natl. Acad. Sci. USA* **102**, 4978 (2005).
- ⁴⁹See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevB.86.045215> for detailed calculations of the sum-frequency susceptibility when the HH-LH splitting is nonzero.
- ⁵⁰L. Fu, C. L. Kane, and E. J. Mele, *Phys. Rev. Lett.* **98**, 106803 (2007).
- ⁵¹B. A. Bernevig, T. L. Hughes, and S.-C. Zhang, *Science* **314**, 1757 (2006).
- ⁵²M. König, S. Wiedmann, C. Brune, A. Roth, H. Buhmann, L. W. Molenkamp, X.-L. Qi, and S.-C. Zhang, *Science* **318**, 766 (2007).
- ⁵³D. Hsieh, D. Qian, L. Wray, Y. Xia, Y. S. Hor, R. J. Cava, and M. Z. Hasan, *Nature (London)* **452**, 970 (2008).
- ⁵⁴X.-L. Qi and S.-C. Zhang, *Phys. Today* **63**, 33 (2010).
- ⁵⁵M. Z. Hasan and C. L. Kane, *Rev. Mod. Phys.* **82**, 3045 (2010).
- ⁵⁶N. Bloembergen, *Nonlinear Optics* (Benjamin, New York, 1965).