Optical characterization of topological insulator surface states: Berry curvature-dependent response

Pavan Hosur
Department of Physics, University of California, Berkeley

We study theoretically the optical response of the surface states of a topological insulator, especially the generation of helicity-dependent direct current by circularly polarized light. Interestingly, the dominant current, due to an interband transition, is controlled by the Berry curvature of the surface bands. This extends the connection between photocurrents and Berry curvature beyond the quasiclassical approximation where it has been shown to hold. Explicit expressions are derived for the (111) surface of the topological insulator Bi$_2$Se$_3$ where we find significant helicity dependent photocurrents when the rotational symmetry of the surface is broken by an in-plane magnetic field or a strain. Moreover, the dominant current grows linearly with time until a scattering occurs, which provides a means for determining the scattering time. The dc spin generated on the surface is also dominated by a linear-in-time, Berry curvature dependent contribution.

I. INTRODUCTION

Topological insulators (TIs) are characterized by topologically protected surface states (SSs). In their simplest incarnation, these correspond to the dispersion of a single Dirac particle, which cannot be realized in a purely two dimensional band structure with time reversal invariance. This dispersion is endowed with the property of spin-momentum locking, i.e., for each momentum there is a unique spin direction of the electron. Most of the experimental focus on TIs so far has been towards trying to directly observe these exotic SSs in real or momentum space, in tunneling and photoemission experiments, respectively, and establish their special topological nature. However, there has so far been a dearth of experiments which study the response of these materials to external perturbations, such as an external electromagnetic field.

In order to fill this gap, we study here the response of TI surfaces to circularly polarized (CP) light. Since photons in CP light have a well-defined angular momentum, CP light can couple to the spin of the surface electrons. Then, because of the spin-momentum-locking feature of the SSs, this coupling can result in dc transport which is sensitive to the helicity (right- vs left-circular polarization) of the incident light. This phenomenon is known as the circular photogalvanic effect (CPGE). In this work, we derive general expressions for the direct current on a TI surface as a result of the CPGE at normal incidence: we look for perturbations that result in a non-zero Berry curvature. Thus, it is exciting that it appears in the response here. Its main implication here is that it gives us a simple rule, in addition to the requirement of the right symmetries, for identifying the perturbations that can give a linear-in-time CPGE at normal incidence; we look for perturbations that result in a non-zero Berry curvature. Put another way, we can identify perturbations that have the right symmetries but still do not give this current because the Berry curvature vanishes for these perturbations. Importantly, for TI SSs, the requirement of a non-zero Berry curvature amounts to the simple physical condition that the spin-direction of the electrons have all three components non-zero. In other words, if the electron spin in the SSs is completely in-plane, the Berry curvature is zero and no linear-in-time CPGE is expected. The spins must somehow be tipped slightly out of the plane, as shown in Fig1a, in order to get such a response. Thus, a pure Dirac (linear) dispersion, for which the spins are planar, cannot give this response; deviations from linearity, such as the hexagonal warping on the (111) surface of Bi$_2$Se$_3$, are essential for tilting the spins out of the plane.

CPGE has been observed in the past in GaAs and SiGe quantum wells - both systems with strong spin-orbit coupling. However, no connection with the Berry curvature was made in those cases. The connection, if present, may be harder to find because the description there necessarily involves transitions between four bands - two spin-orbit split valence bands and two spin-
We find that a current density of $\sim 100nA/mm$ ($\sim 10nA/mm$) can be obtained due to the CPGE with a 1Watt laser. This value can be easily measured by current experimental techniques. Conversely, the scattering time, crucial for transport processes, for Bi$_2$Se$_3$ SSs can be determined by measuring the current. In comparison, circular photogalvanic currents of a few nanoamperes per Watt of laser power have been measured in GaAs and SiGe quantum wells.

A connection between the optical response of a system and the Berry curvature of its bands has been previously noted at the low frequencies, where a semiclassical mechanism involving the anomalous velocity of electrons in a single band explains it\textsuperscript{[14]}. Here, we show it for inter band transitions where no quasiclassical approximation is applicable. Instead, we calculate the quadratic response function directly. A connection is still present which points to a deeper relation between the response functions and the Berry curvature.

This paper is organized as follows. In Sec. II we state the symmetry conditions under which a CPGE may occur. We present our results, both general as well as for Bi$_2$Se$_3$ in particular, in Sec. III A and describe the microscopic mechanism in Sec. III B. The calculation is described briefly in Sec. III C and in detail in Appendix B. In Sec. IV we give our results for dc spin.

\section{Symmetry Considerations for the CPGE}

In this section, we specify the symmetry conditions under which one can get a CPGE on the surface of a TI. But first, let us briefly review the concept of the CPGE in general.

The dominant dc response of matter to an oscillating electric field is, in general, quadratic in the electric field. When the response of interest is a current, the effect is known as the photogalvanic effect. This current can be written as

$$j_\alpha = \eta_{\alpha\beta\gamma}e_{\beta}(\omega)e_{\gamma}(-\omega)$$  \hspace{1cm} (1)

where $e_{\alpha}(t) = e_{\alpha}(\omega)e^{i\omega t} + e_{\alpha}^*(\omega)e^{-i\omega t}$ is the incident electric field, $e_{\alpha}^*(\omega) = e_{\alpha}(-\omega)$ and $\eta_{\alpha\beta\gamma}$ is a third rank tensor, which has non-zero components only for systems that break inversion symmetry, such as the surface of a crystal.

For $j_\alpha$ to be real, one has $\eta_{\alpha\beta\gamma} = \eta_{\gamma\alpha\beta}^*$. Thus, the real (imaginary) part of $\eta_{\alpha\beta\gamma}$ is symmetric (anti-symmetric) under interchange of $\beta$ and $\gamma$, and therefore describes a current that is even (odd) under the transformation $\omega \rightarrow -\omega$. Consequently, $j_\alpha$ can be conveniently sepa-
rated according to
\[ j_α = S_{αβγ} \left( \mathcal{E}_β(ω)\mathcal{E}_γ^*(ω) + \mathcal{E}_γ(ω)\mathcal{E}_β^*(ω) \right) + i\Lambda_{αμ}(\mathcal{E} \times \mathcal{E}^*)μ \]

(2)

where \( S_{αβγ} \) is the symmetric part of \( η_{αβγ} \) and \( Λ_{αμ} \) is a second-rank pseudo-tensor composed of the anti-symmetric part of \( η_{αβγ} \). For CP light, \( \mathcal{E} \propto \hat{x} ± i\hat{y} \) if \( \hat{z} \) is the propagation direction and only the second term in Eq. (2) survives, and hence represents the CPGE. This effect is odd in \( ω \). On the other hand, the first term, which is even in \( ω \), represents the linear photogalvanic effect as it is the only contribution for linearly polarized light. Since the transformation \( ω \to −ω \), or equivalently, \( \mathcal{E} \to \mathcal{E}^* \) reverses the helicity of CP light, i.e., changes right-CP light to left-CP light and vice versa, the CPGE is the helicity-dependent part of the photogalvanic effect.

The helicity of CP light is odd (i.e., right- and left-CP light get interchanged) under time-reversal. It is also odd under mirror reflection about a plane that contains the incident beam, but invariant under arbitrary rotation about the direction of propagation. Let us consider normal incidence of CP light on a TI surface normal to the \( z \) axis. Let us further assume that there is a mirror symmetry \( R_z \) about the \( z \)-axis (such as the threefold rotation symmetry on the (111) surface of \( Bi_2Se_3 \)), then no surface helicity-dependent direct photocurrent is permitted. One needs to break this rotation symmetry completely by, for example, and in-plane magnetic field, strain etc., to obtain a nonvanishing current.

III. HELICITY-DEPENDENT DIRECT PHOTOCURRENT

We now present our main results for the photocurrent and estimate it for \( Bi_2Se_3 \). After painting a simple microscopic picture for the mechanism, we give a brief outline of the full quantum mechanical treatment of the phenomenon.

A. Results

A general two-band Hamiltonian (in the absence of the incident light) can be written as

\[ \hat{H} = \sum_\mathbf{p} H_\mathbf{p} = \sum_\mathbf{p} |E_\mathbf{p}| \hat{\mathbf{n}}(\mathbf{p}) σ \]

(3)

upto a term proportional to the identity matrix, which is not important for our main result which involves only inter-band transitions. Here \( \hat{\mathbf{n}}(\mathbf{p}) \) is a unit vector and \( σ \) are the spin-Pauli matrices. Clearly, this can capture a Dirac dispersion, eg. with \( E(\mathbf{p}) = ±v_F p \) and \( \hat{\mathbf{n}}(\mathbf{p}) = v_F \hat{z} \times \mathbf{p} \). It can also capture the SSs of \( Bi_2Se_3 \) in the vicinity of the Dirac point, which includes deviations beyond the Dirac limit. We also assume the Hamiltonian has a reflection symmetry \( m \) about \( y \)-axis, where \( \hat{z} \) is the surface normal. Using the zero temperature quadratic response theory described in Sec III C, we calculate the current due to the CPGE and find that

\[ j_{CPGE}(t) = (j_{a1} + j_{a2}(t)) \hat{x} \]

(4)

where the subscripts \( a, (na) \) stand for “absorptive” and “non-absorptive”, respectively. The absorptive part of the response involves a zero momentum interband transition between a pair of levels separated by energy \( hω \). These terms are only non zero when there is one occupied and one empty level. In this part of the response, we find a term that is time-dependent, \( j_{a2}(t) \). In particular, this term grows linearly with the time over which the electromagnetic perturbation is present, which is allowed for a dc response. In reality, this linear growth is cut off by a decay process which equilibrates populations, and is characterized by a time constant \( τ \). In clean samples at sufficiently low temperatures, characterized by large \( τ \), this contribution is expected to dominate the response, and hence, is the focus of our work. The other contributions are discussed in Appendix B. Conversely, because of the linear growth with time, one can determine the lifetime of the excited states by measuring the photocurrent. This term is

\[ j_{a2}(t) = -\frac{πe^3}{4} \frac{ℏ^2 sgn(ω)}{ω} \sum_\mathbf{p} δ(|hω| − 2|E_\mathbf{p}|) v_x(\mathbf{p}) F(\mathbf{p}) \]

(5)

where we have assumed that the chemical potential is in between the two energy levels \( ±|E_\mathbf{p}| \) connected by the optical frequency \( hω \), and that temperature can be neglected compared to this energy scale. Here, \( v_x(\mathbf{p}) = \frac{∂E_\mathbf{p}}{∂p_x} \) is the conventional velocity and \( F(\mathbf{p}) = i\sum_\mathbf{p} (|∂_p u(\mathbf{p})| |∂_p u(\mathbf{p})| + c.c.) \), where \( |u(\mathbf{p})| \) is the conduction band Bloch state at momentum \( \mathbf{p} \). is the Berry curvature of the conduction band at momentum \( \mathbf{p} \). For the class of Hamiltonians \( \mathcal{H} \) that we are concerned with, the Berry curvature is given by (See Appendix A):

\[ F(\mathbf{p}) = \hat{\mathbf{n}} \left( \frac{∂\hat{\mathbf{n}}}{∂p_x} × \frac{∂\hat{\mathbf{n}}}{∂p_y} \right) \]

(6)

which is the skyrmion density of the unit vector \( \hat{\mathbf{n}} \) in momentum space. Since \( ∂_p \hat{\mathbf{n}} \perp \hat{\mathbf{n}} \) for \( i = x, y, F(\mathbf{p}) \neq 0 \) only if all three components of \( \hat{\mathbf{n}} \) are nonvanishing. For linearly dispersing bands, \( \hat{\mathbf{n}} \) has only two non-zero components (eg. \( H_p = p_y σ_y - p_x σ_y, \hat{\mathbf{n}} \propto (p_y, -p_x, 0) \)). Hence, corrections beyond the pure Dirac dispersion are essential. Also, due to \( m \), the Berry curvature satisfies \( F(p_x, p_y) = -F(−p_x, p_y) \). Since in Eq. (5) we have multiplied the Berry curvature, which also
transforms the same way, a finite contribution is obtained on doing the momentum sum.

We now calculate $j_{a2}(t)$ for the threefold-symmetric (111) surface of Bi$_2$Se$_3$ starting from the effective Hamiltonian $^8$

$$H = v_F (p_x \sigma_y - p_y \sigma_x) + \frac{\lambda}{2}(p_x^2 + p_y^2) \sigma_z \quad (7)$$

where $v_F \approx 5 \times 10^5 m/s$ $^{12}$ and $\lambda = 50.1 eV \cdot Å^3$. A spin independent quadratic term has been dropped since it does not modify the answers for interband transitions, which only involve the energy difference between the bands.

To get a non-zero $j_{CPGE}$, the threefold rotational symmetry must be broken, which we first propose to do by applying a magnetic field $B$ in the $x$-direction. This field has no orbital effect, and can be treated by adding a Zeeman term $-g_\mu_B B \sigma_x$, where $g_\mu$ is the appropriate g-factor and $\mu_B$ is the Bohr magneton, to the Hamiltonian $^7$. To lowest order in $\lambda$ and $B$, we get

$$j_{a2}(t) = \frac{3e^3 v_F \xi_2^2 (g_\mu B)}{16 \hbar^2} A \left( \frac{\lambda B}{2} \right)^2 t \quad (8)$$

to lowest order in $\lambda$ and $B^2$, where $A$ is the laser spot-size. For $g_\mu = 0.5$, and assuming the experiment is done in a $10T$ field with a continuous wave laser with $h\omega = 0.1 eV$ which is less than the bulk band gap of $0.35 eV$, $A \sim 1 mm^2$, a laser power of $1 W$, and the spin relaxation time $t \sim 10 ps$, we get a current density of $\sim 100 nA/mm$, which is easily measurable by current experimental techniques. Note that the expression (8) for $j_{a2}(t)$ contains the parameter $\lambda$ which measures the coupling to $\sigma_z$ in Eq. (7). Since $B = B\hat{x}$ breaks the rotation symmetry of the surface completely, a naive symmetry analysis suggests, wrongly, that deviations from linearity, measured by $\lambda$, are not needed to get $j_{a2}(t)$.

The rotation symmetry can also be broken by applying a strain along $x$, which can be modeled by adding a term $\delta \lambda p_x^2 \sigma_z$ to $H$ in Eq. (7). This gives

$$j_{a2}(t) = \frac{3e^3 v_F (\delta \lambda) \xi_2^2 \omega t}{2} A \quad (9)$$

to lowest order in $\lambda$ and $\delta \lambda$. For a 1% strain, $\delta \lambda/\lambda = 0.01$, and the same values for the other parameters as in Eq. (8), we get a current density of $\sim 100 nA/mm$. Eq. (9) does not contain $\lambda$; this is because $\delta \lambda$ alone both breaks the rotation symmetry and tips the spins out of the $xy$-plane.

B. Physical process

The appearance of the Berry curvature suggests a role of the anomalous velocity in generating the current. Such mechanisms have been discussed in the literature in the context of the CPGE $^{14, 16}$. However, those mechanisms only work when the electric field changes slowly compared to the typical scattering time. The SSs of Bi$_2$Se$_3$ probably have lifetimes of tens of picoseconds, and thus, we are in the opposite limit when $\hbar \omega = 0.1 eV$, which corresponds to a time scale $10^3$ times shorter.

In this limit, the dc responses are a result of a preferential absorption of the photon at one of the two momentum points for each pair of points $(\pm p_x, p_y)$ related by $m$, as shown in Fig. 1 for $p_y = 0$. According to the surface Hamiltonian $^7$, the spin vector $S = \mp \hbar \hat{z}$ gets tipped out of the $xy$-plane for states that lie beyond the linear dispersion regime, but the direction of the tipping is opposite for $(p_x, p_y)$ and $(-p_x, p_y)$. Thus, photons of helicity $-1$, which can only raise $\langle S_z \rangle$ of an electron, are preferably absorbed by the electrons that have $\langle S_z \rangle < 0$ in the ground state. The response, then, is determined by the properties of these electrons. Clearly, the process is helicity-dependent as reversing the helicity would cause electrons with $\langle S_z \rangle > 0$ to absorb the light preferentially.

This is consistent with the requirement of a non-zero Berry curvature, which essentially amounts to the spin direction $\hat{n}$ having to be a three-dimensional vector. In the linear limit, where $H = v_F (p_x \sigma_y - p_y \sigma_x)$, the spin is entirely in-plane, and all the electrons absorb the incident light equally.

C. Calculation in brief

We now briefly outline the calculation of the helicity-dependent photocurrent. The detailed calculation can be found in Appendix B. Readers only interested in our results may wish to skip this section.

The Model: The Hamiltonian and relevant electric field (vector potential) perturbations for getting a direct current to second order in the electric field of the incident photon are

$$H = |E_p| \hat{n}(p) \cdot \sigma \quad (10)$$

$$H' = j_x A_x(t) + j_y A_y(t) \quad (11)$$

$$j_x = \frac{\partial H'}{\partial p_0} \quad (12)$$

$$A_x(t) + i A_y(t) = A_0 e^{i(\omega - i \epsilon)t} \quad (13)$$

where $A$ is the vector potential, $\hat{z}$ is assumed to be the surface normal, and $\epsilon$ is a small positive number which ensures slow switch-on of the light.

Quadratic response Theory: In general, the current along $x$ to all orders in the perturbation $H'$ is

$$\langle j_x \rangle(t) = \left\langle T^* \left( e^{i \int_{-\infty}^t dt' H'(t') \right) j_x(t) T \left( e^{-i \int_{-\infty}^t dt' H'(t') \right) \right) \right\rangle \quad (14)$$

where $T(T^*)$ denotes time-ordering (anti-time-ordering) and $O(t) = e^{iHt} O e^{-iHt}$. Terms first order in $H'$ cannot give a direct current. The contribution to the current
from the second order terms can be written as
\[
\langle j_x(t) \rangle = \int_{-\infty}^{\infty} dt' \int_{-\infty}^{t_1} dt'' \langle [j_x(t'), H'(t''), H'] \rangle
\]
\[
= \int_{-\infty}^{t} dt' \int_{-\infty}^{t_1} dt'' \chi_{x \alpha \beta}(t, t', t'') A_{\alpha}(t') A_{\beta}(t'')
\]
\[
= \sum_p \frac{1}{2} \text{Tr} \left\{ \left( 1 - \frac{H}{|E_p|} \right) O \right\} = - \sum_p \frac{\text{Tr}(HO)}{2|E_p|}
\]
\[
\langle O \rangle = \sum_p \frac{1}{2} \text{Tr} \left\{ \left( 1 - \frac{H}{|E_p|} \right) O \right\} = - \sum_p \frac{\text{Tr}(HO)}{2|E_p|}
\]
\[
\chi_{x \alpha \beta}(t_1, t_2) = - \sum_p \frac{\text{Tr}(H[\hat{j}_x, l_\alpha(t_1) \cdot l_\beta(t_2)])}{2|E_p|}
\]
Eq. (17) is the zero temperature limit of the finite temperature expression for the quadratic susceptibility proven in Ref. [7].

Because of the mirror symmetry $m$, $\chi_{x \alpha \beta}(t_1, t_2)$ is non-vanishing only for $\alpha \neq \beta$. To get a direct current, we retain only the non-oscillating part of $A_{x}(t+t_1)A_{y}(t+t_2) = \frac{A^2}{2}e^{2\omega t} \{ \sin\left(2\omega t + \omega(t_1 + t_2)\right) - \sin\left(\omega(t_1 - t_2)\right) \}$. Thus,
\[
j_{x}^{dc}(t) = \frac{A^2 e^{2\omega t}}{4} \int_{-\infty}^{0} dt_1 \int_{-\infty}^{t_1} dt_2 \left\{ (\chi_{xy} - \chi_{yx})(t_1, t_2) \times e^{(t_1+t_2)} \sin (\omega(t_2 - t_1)) \right\}
\]
\[
\textbf{The Result:} \text{ After carrying out the two time-integrals, we get the three currents mentioned in Eq. [4]. For clean samples at low temperatures, } j_{a2}(t), \text{ which grows linearly with time, is expected to dominate. A general expression for this term is (in the units } e = h = v_F = 1 \text{ where } v_F \text{ is the Fermi velocity) }
\]
\[
j_{a2}(t) = \frac{i A^2 \pi \text{sgn}(\omega)}{2\omega^2} \sum_p \delta(|\omega| - 2|E_p|) \text{Tr}(Hj_x) \text{Tr}(H[j_x, j_y])
\]
Using Eqs. (10) and (12) and the Lie algebra of the Pauli matrices, $[\sigma_i, \sigma_j] = 2i \epsilon_{ijk} \sigma_k$ where $\epsilon_{ijk}$ is the anti-symmetric tensor, the above traces can be written as
\[
\text{Tr}(Hj_x) = 2|E_p| v_x(p)
\]
\[
\text{Tr}(H[j_x, j_y]) = 4i|E_p|^3 \hat{n} \left( \frac{\partial \hat{n}}{\partial p_x} \times \frac{\partial \hat{n}}{\partial p_y} \right)
\]
\[
= 4i|E_p|^3 F(p)
\]
Eqs. (10), (20) and (21) give our main result Eq. (5).

\section{IV. SPIN GENERATION}

Having understood the microscopic mechanism underlying the generation of the photocurrent $j_{a2}(t)$, we wonder, next, whether such a population imbalance can lead to any other helicity-dependent macroscopic responses. Since each absorbed photon flips the $z$-component of the spin of an electron, a net $\langle S_z \rangle$ is expected to be generated on the surface.

The calculation of $\langle S_z \rangle$ is identical to that of $j_{CPGE}$. The total $\langle S_z \rangle$ generated consists of the same three parts as $j_{CPGE}$, and the dominant part is
\[
S_{a2}^z(t) = \frac{-e^2 E_0^2 \hbar \text{sgn}(\omega)}{8} \sum_p \delta(|\omega| - 2|E_p|) n_z(p) F(p)
\]
$S_z$ does not break the rotational symmetry of the surface, so we calculate $S_{a2}^z(t)$ directly for the threefold symmetric Hamiltonian (7) and obtain
\[
S_{a2}^z(t) = \frac{e^2 E_0^2 (\hbar \omega)^3 \lambda_2 t}{210} \mathcal{A}
\]
For the same values of all the parameters as for $j_{a2}(t)$, we get $S_{a2}^z(t) \sim 10h$, which means only ten electron spins are flipped over an area of $\sim 1mm^2$. If, instead, we ignore the cubic corrections but assume magnetic ordering on the surface, so that $H = v_F(p_x \sigma_y - p_y \sigma_x) + M \sigma_z$, we get
\[
S_{a2}^z(t) = \frac{-e^2 E_0^2 \lambda_2 M^2 t}{10(\hbar \omega)^3} \mathcal{A}
\]
which again gives a rather small value of $\sim 10h$ for $M \sim 10K$, a typical magnetic ordering transition temperature. However, the spin generated could be measurable if one uses a pulsed laser of, say, MegaWatt power, and performs a time-resolved experiment.

\section{V. CONCLUSIONS}

In summary, we studied the CPGE on the surface of a TI at normal incidence, and applied the results to the (111) surface of Bi$_2$Se$_3$. If the rotational symmetry of the TI surface is broken by applying an in-plane magnetic field or a strain, we predict an experimentally measurable direct photocurrent. A striking feature of this current is that it depends on the Berry curvature of the
electron bands. Such a dependence can be understood intuitively as a result of the incident photons getting absorbed unequally by electrons of different momenta and hence, different average spins. The current grows linearly with time until a decay process equilibrates populations, which provides a way of determining the excited states lifetime. We also calculated the amount of dc helicity-dependent out-of-plane component of the electron spin generated. This does not require any rotational symmetry breaking; however, the numerical value is rather small with typical values of parameters.

In the future, we hope to find a generalization of our results for oblique incidence. Experimentally, this is a very attractive way of breaking the rotational symmetry of the surface; indeed, such experiments have already been performed successfully on graphene [19]. In graphene, helicity-dependent direct photocurrents have also been predicted by applying a dc bias [13]. However, with a dc bias across a TI surface and ordinary continuous lasers, we find the current to be too low to be measurable. Finally, we also wonder whether the Berry curvature dependence of the helicity-dependent response to CP light survives for three- and higher-band models. If it does, it would be interesting to write such a model for semiconductor quantum wells such as GaAs and SiGe. It could also enable one to treat oblique incidence, by considering transitions to higher bands of different parities, because they are driven by the normal component of the electric field, $E_z$.

We would like to thank Ashvin Vishwanath for enlightening discussions, Joseph Orenstein for useful experimental inputs, and Ashvin Vishwanath and Yi Zhang for invaluable feedback on the draft.

This work was supported by LBNL DOE-504108.

**Appendix A: Proof of Berry curvature expression**

Here we show that the Berry curvature defined for Bloch electrons as

$$F(p) = i \left( (\partial_{p_x} u)(\partial_{p_y} u) - (\partial_{p_y} u)(\partial_{p_x} u) \right)$$  \hspace{1cm} (A1)

can be written as

$$F(p) = \mathbf{n}(p) \cdot \left( \partial_{p_x} \mathbf{n} \times \partial_{p_y} \mathbf{n} \right)$$  \hspace{1cm} (A2)

for the band with energy $|E_p|$ for Hamiltonians of the form $H_p = |E_p| \mathbf{n}(p) \cdot \mathbf{\sigma}$.

At momentum $p$, the Bloch state $|u_p\rangle$ with energy $|E_p|$ is defined as the state whose spin is along $\mathbf{n}(p)$. Defining $|\uparrow\rangle$ as the state whose spin is along $+z$, $|u_p\rangle$ is obtained by performing the appropriate rotations,

$$|u_p\rangle = e^{-i \theta(p)} \phi(p) e^{i \phi(p)} |\uparrow\rangle$$  \hspace{1cm} (A3)

where $\theta(p)$ and $\phi(p)$ are the polar angles that define $\mathbf{n}(p)$:

$$\mathbf{n}(p) = \sin \theta(p) \cos \phi(p) \mathbf{x} + \sin \theta(p) \sin \phi(p) \mathbf{y} + \cos \theta(p) \mathbf{z}$$  \hspace{1cm} (A4)

Substituting Eq. (A3) in Eq. (A1), one gets

$$F(p) = \sin \theta(p) \left( \partial_{p_x} \theta(p) \phi(p) - \partial_{p_y} \phi(p) \partial_{p_x} \theta(p) \right)$$  \hspace{1cm} (A5)

which, on using Eq. (A4) and some algebra, reduces to the required expression Eq. (A2).

**Appendix B: current calculation for the cpge**

Here we explain the current-calculation of Sec. IIIA in more detail and also state results for the parts of the current that we chose not to focus on there.

As shown in Sec. III C the relevant susceptibility is

$$\chi^{\alpha\beta}(t, t', t'') = -\frac{1}{2} \sum_p \text{Tr} \left( \frac{H}{|E_p|} \left[ j^x(t), j^\alpha(t') \right], j^\beta(t'') \right)$$

$$= -\sum_p \frac{1}{2|E_p|} \text{Tr} \left( H \left[ [j^x, j^\alpha(t_1)], j^\beta(t_2) \right] \right)$$

$$\equiv \chi^{\alpha\beta}(t_1, t_2)$$  \hspace{1cm} (B1)

where $t_1 = t' - t$, $t_2 = t'' - t$, and the non-vanishing components of $\chi^{\alpha\beta}$ are those for which $\alpha \neq \beta$. The non-oscillating part of the current, hence, is

$$\langle j^{de}_x (t) \rangle = j_{CPGE}^x(t) = \frac{A_0 e^{2et}}{4} \int_{-\infty}^{t_1} dt_1 \int_{-\infty}^{t_2} dt_2$$

$$\left( \chi^{xx}(t_1, t_2) - \chi^{xy}(t_1, t_2) \right) e^{i(t_1 + t_2)} \sin(\omega(t_2 - t_1))$$  \hspace{1cm} (B2)

Since $j_{CPGE}^x(t)$ is an odd function of $\omega$, it reverses on reversing the polarization, as expected.

The traces in the susceptibility expressions are calculated by introducing a complete set of states in place of the identity several times. Thus,

$$\chi^{xx}(t_1, t_2)$$  \hspace{1cm} (B3)

$$= -\sum_p \frac{1}{2|E_p|} \text{Tr} \left( H \left[ [j^x, j^x(t_1)], j^y(t_2) \right] \right)$$

$$= -\sum_p \sum_{nml} \text{sgn}(E_n) \left\{ e^{i(E_m - E_n)t_2} \times \right.$$  \hspace{1cm} (B4)

$$e^{i(E_l - E_m)t_1} - e^{-i(E_l - E_m)t_1} \right\} X_{nl} X_{lm} Y_{mn} + \text{c.c.} \right\}

where $X_{nl} = \langle n | j_x | m \rangle$ etc. and the subscript $p$ on $E_p$ has been dropped to enhance the readability. Similarly,

$$\chi^{xy}(t_1, t_2)$$

$$= -\sum_p \frac{1}{2|E_p|} \text{Tr} \left( H \left[ [j^y, j^x(t_1)], j^x(t_2) \right] \right)$$

$$= -\sum_p \sum_{nml} \text{sgn}(E_n) \left\{ e^{i(E_m - E_n)t_2} X_{mn} \times \right.$$  \hspace{1cm} (B5)

$$e^{i(E_l - E_m)t_1} X_{nl} Y_{lm} - e^{-i(E_l - E_m)t_1} Y_{nl} X_{lm} \right\} + \text{c.c.} \right\}$
Substituting (B3) and (B4) in (??), we get

\[ j_{CPE}(t) = \frac{A_0^2 e^{2\epsilon t}}{4} \text{Re} \int_0^{t_1} dt_1 \int_{-\infty}^{t_1} dt_2 e^{i(\epsilon t_1 + t_2)} \times \]

\[ \sin(\omega(t_1 - t_2)) \sum_{p, n, m l} \text{sgn}(E_n) e^{i(E_m - E_n) t_2} \times \]

\[ \left\{ e^{i(E_l - E_m) t_2} - e^{-i(E_l - E_m) t_2} \right\} X_{nl} X_{lm} Y_{mn} - \]

\[ X_{mn} \left( e^{i(E_l - E_m) t_1} X_{nl} Y_{mn} - e^{-i(E_l - E_m) t_1} Y_{nl} X_{lm} \right) \}

where \( \text{Re} \) stands for ‘the real part of’. Carrying out the the two time integrations gives

\[ j_{CPE}(t) = \frac{A_0^2 e^{2\epsilon t}}{8} \text{Im} \sum_p \sum_{n, m l} \text{sgn}(E_n) \times \]

\[ \left[ \frac{1}{E_m - E_n + \omega - i\epsilon} - \frac{1}{E_m - E_n - \omega - i\epsilon} \right] \times \]

\[ \left\{ X_{nl} (X_{lm} Y_{mn} - Y_{lm} X_{mn}) + X_{ml} (Y_{mn} X_{nl} - X_{mn} Y_{nl}) \right\} \]

where \( \text{Im} \left( \frac{1}{\Omega - i\epsilon} \right) = \pi \delta(\Omega) \) and \( \text{Re} \left( \frac{1}{\Omega - i\epsilon} \right) = \frac{1}{\Omega} \) in the limit \( \epsilon \to 0 \), we get after some algebra, \( j_{CPE}(t) = j_{na} + j_{a1} + j_{a2}(t) \), where \((\text{Tr} \) denotes the trace) \( \text{Tr}(H j_x) \text{Tr}(H [j_x, j_y]) \)

\[ j_{a1} = - \frac{\pi A_0^2 \text{sgn}(\omega)}{32} \sum_p \delta(|\omega| - 2|E_p|) \times \]

\[ \text{Tr}(H [j_x, j_y]) \]

\[ \frac{E_p^2}{|E_p|^2} \]

which also results from interband absorption and increases linearly in time. The last term was the main focus of our work.

[1] P. Roushan et al., Nature 460, 1106-1109 (2009).
[2] D. Hsieh, D. Qian, L. Wray, Y. Xia, Y. Hor, R. J. Cava, and M. Z. Hasan, Nature 452, 970 (2008).
[3] Y. Xia, L. Wray, D. Qian, D. Hsieh, A. Pal, H. Lin, A. Bansil, D. Grauer, Y. S. Hor, R. J. Cava, M. Z. Hasan, Nature Physics Vol. 5, No. 6, pp398 (2009)
[4] F. D. M. Haldane, Phys. Rev. Lett. 93, 206602 (2004).
[5] Thouless, Kohmoto, Nightingale, den Nijs, Phys. Rev. Lett. 49, 405, (1982).
[6] Ganichev et al., PRL 86, 4358 (2001).
[7] Ganichev et. al., Mat. Res. Soc. Symp. Proc. Vol. 690, F3.11.1 (2002).
[8] Chao-Xing Liu, Xiao-Liang Qi, HaiJun Zhang, Xi Dai, Zhong Fang, Shou-Cheng Zhang, [arXiv:1005.1682]
[9] Y. L. Chen, J. G. Analytis, J. H. Chu, Z. K. Liu, S. K. Mo, X. L. Qi, H. J. Zhang, D. H. Lu, X. Dai, Z. Fang, S. C. Zhang, I. R. Fisher, Z. Hussain, Z. X. Shen, Science Vol. 325 no. 5937, pp178 (2009).
[10] L. Fu, Phys. Rev. Lett. 103, 266801 (2009).
[11] D. Hsieh, Y. Xia, D. Qian, L. Wray, J. H. Dil, F. Meier, J. Osterwalder, L. Patthey, J. G. Checkelsky, N. P. Ong, A. V. Fedorov, H. Lin, A. Bansil, D. Grauer, Y. S. Hor, R. J. Cava & M. Z. Hasan, Nature 460, 1101 (2009).
[12] Haijun Zhang, Chao-Xing Liu, Xiao-Liang Qi, Xi Dai, Zhong Fang & Shou-Cheng Zhang, Nature Physics 5, 438-442 (2009).
[13] Munoz, Perez, Vina, Ploog, Phys. Rev. B 51, 4247 (1995).
[14] E. Deyo et al., [arXiv:0904.1917v1]
[15] Ong, Lee, Foundations of Quantum Mechanics, ed. Sachio Ishioka and Kazuo Fujikawa (World Scientific, 2006), p. 121.
[16] J. E. Moore, J. Orenstein, [arXiv:0911.3630v1]
[17] Ch. 7, ‘Nonlinear Optical Phenomena’, Paul N. Butcher, Eq. 7.25 and preceding discussion.
[18] Oka, Aoki, Phys. Rev. B 79, 081406(R), 2009.
[19] Karch et al., [arXiv:1002.1047v1]