Detection of quantum geometric tensor by nonlinear optical response

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(Dated: August 11, 2021)

Quantum geometric tensor (QGT), including a symmetric real part defined as quantum metric and an antisymmetric part defined as Berry curvature, is essential for understanding many phenomena. We studied the photogalvanic effect of a multiple-band system with time-reversal-invariant symmetry by theoretical analysis in this work. We concluded that the integral of gradient of the symmetric part of QGT in momentum space is related to the linearly photogalvanic effect, while the integral of gradient of Berry curvature is related to the circularly photogalvanic effect. Our work offered an alternative interpretation for the photogalvanic effect in the view of QGT, and a simple approach to detect the QGT by nonlinear optical response.

PACS numbers:

Introduction

Geometry plays an important role in many aspects of modern physics. The geometry of the eigenstates is encoded in the quantum geometric tensor (QGT), which is defined on any manifold of the eigenstates. Knowledge of the quantum metric defines the distance between nearby states. The Berry curvature is crucial for topological phenomena, such as, orbital magnetic susceptibility, the exciton Lamb shift, and anomalous Hall drift. The nonlinear optical (NLO) response, including second harmonic generation (SHG) and photogalvanic effect, has wide applications in scientific community.

For example, SHG is used for frequency doubling of laser light, and detection of the breaking of spatial inversion symmetry. The photogalvanic effect (PGE) is determined by the helicity of light, and CPGE can be used for the detection of topological charge of quantum matter. The direction of shift current is independent from the helicity of light, and usually dubbed as linear photogalvanic effect (LPGE). Recently, the Berry curvature dipole (BCD) defined as the integral of the gradient of the Berry curvature in momentum space, affords a new interpretation for PGE. Bilayer WTe$_2$ with tilted Weyl point provides a possible platform to detect the BCD and its induced nonlinear Hall current. The nonlinear Hall effect induced by BCD implies that the nonlinear response may have some underlying connection with the QGT. We may wonder that what is the role of symmetric real part of QGT for nonlinear optical response, and can we detect QGT by nonlinear optical response?

In this letter, we address the basic theory of GPE of multiple-band system with time-reversal-invariant symmetry (TRIS). We concluded that the gradient of symmetric real part of QGT is related to the LPGE, while the gradient of Berry curvature is related to the CPGE. Our work builds close connections between nonlinear optical response of a system with time-reversal-invariant symmetry and the geometry of quantum states, and facilitates the detection of QGT by nonlinear optical response.

Dynamics of density matrix

Under spatial homogeneous external field $\vec{E}(t) = \int_0^\infty d\omega [\bar{E}(\omega)e^{-i\omega t} + c.c.]$, the light-matter interaction can be described by below model Hamiltonian,

$$\hat{H}(t) = \int d\vec{r} \Psi^\dagger(\vec{r}, t)[\hat{h}_0 - \epsilon \vec{E}(\vec{r}) \cdot \vec{r}]\Psi(\vec{r}, t),$$

where $\hat{h}_0$ is the unperturbed Hamiltonian describing the ground state. The orthogonal Bloch functions $\phi_n(\vec{k}, \vec{r})$ satisfy

$$\hat{h}_0 \phi_n(\vec{k}, \vec{r}) = \epsilon_n(\vec{k}) \phi_n(\vec{k}, \vec{r}),$$
in which \( n \) is the band index and \( \vec{k} \) is position in momentum space. The Bloch functions are orthogonal to each other,

\[
\int d\vec{r}' \phi^*_n(\vec{k}, \vec{r}) \phi_m(\vec{k}', \vec{r}') = \delta_{nm} \delta(\vec{k} - \vec{k}').
\] (4)

Wave function \( \Psi(\vec{r}, t) \) can be expressed as combination of Bloch functions with annihilation operators \( a_n(\vec{k}) \),

\[
\Psi(\vec{r}, t) = \sum_n \int_{BZ} \frac{d^3\vec{k}}{(2\pi)^3} a_n(\vec{k}, t) \phi_n(\vec{k}, \vec{r}) = \sum_n a_n(\vec{k}, t) \phi_n(\vec{k}, \vec{r}).
\] (5)

The velocity operator \( \frac{i}{\hbar} [\hat{H}, \phi_n(\vec{k}, \vec{r})] \) is defined as

\[
\vec{v} = \frac{i}{\hbar} \left[ \hat{H}, \phi_n(\vec{k}, \vec{r}) \right] = \frac{i}{\hbar} \left[ \hat{H}_0, \phi_n(\vec{k}, \vec{r}) \right] - i \frac{e}{\hbar} [ E_n(t), \phi_n(\vec{k}, \vec{r})],
\] (6)

where we assume that summation is performed on repeated index, and the current is expressed as

\[
J_n(t) = \frac{ie}{\hbar} \Psi^*(t) [ \hat{H}, \phi_n ] \Psi(t) = \frac{ie}{\hbar} \int d\vec{k} \partial_\alpha \epsilon_n(\vec{k}) \rho_{nn}(\vec{k}, t) + \frac{e^2 E_n(t)}{\hbar} \int d\vec{k} f^u_{nn}(\vec{k}) \rho_{nm}(\vec{k}, t) + \frac{e^2 E_n(t)}{\hbar} \int d\vec{k} g^u_{nn}(\vec{k}, t)
\] (7)

where \( \partial_\alpha = \frac{\partial}{\partial k_\alpha} \), energy difference \( \epsilon_{nm}(\vec{k}) = \epsilon_n(\vec{k}) - \epsilon_m(\vec{k}) \), and density matrix \( \rho_{nm}(\vec{k}, t) = a^*_n(\vec{k}, t) a_m(\vec{k}, t) \). Here, we used the position matrix \( \frac{\hbar}{2}\text{tr} \) and the definition of Berry connection,

\[
\langle n' \vec{k}' | n \vec{k} \rangle = \int d\vec{r} \phi^*_n(\vec{k}', \vec{r}) \phi_n(\vec{k}, \vec{r}) = A_{n' n}(\vec{k}) \delta(\vec{k} - \vec{k}') - i \delta_{n' n} \nabla_\vec{k} \phi_n(\vec{k}, \vec{r}),
\] (8)

where Berry connection \( A_{n' n}(\vec{k}) = i \langle u_n(\vec{k}) | \nabla_\vec{k} u_{n'}(\vec{k}) \rangle \), in which \( u_n(\vec{k}) = e^{-i\vec{k} \cdot \vec{r}} \phi_n(\vec{k}, \vec{r}) \) is the periodic part of Bloch function. The (static) non-abelian Berry curvature \( f^u_{nn}(\vec{k}) \) is defined as \( [ A_{nm}^u(\vec{k}) \partial_\alpha A^u_{nm}(\vec{k}) ] - i \sum_l [ A_{n'm}^u(\vec{k}) A^u_{im}(\vec{k}) - A_{nm}^u(\vec{k}) A^u_{i'm}(\vec{k}) ] \)

\[
(9)
\]

which is vanishing, and the dynamical Berry curvature \( g^u_{nn}(\vec{k}, t) \) is defined as,

\[
g^u_{nn}(\vec{k}, t) = i \partial_\alpha a^*_n(\vec{k}, t) \partial_\beta a_n(\vec{k}, t) - \partial_\alpha a^*_n(\vec{k}, t) \partial_\beta a_n(\vec{k}, t).
\] (10)

TRIS required that \( g^u_{nn}(\vec{k}, t) = -g^u_{nn}(-\vec{k}, t) \), therefore the dynamical Berry curvature will not cause GPE. Therefore, only the second line and third line may contribute to the second order response.

The dynamics of density matrix \( \rho(\vec{k}, t) \) can be described by the Heisenberg’s equation of motion \( [42] \),

\[
i \hbar \frac{d\rho_{nm}(\vec{k}, t)}{dt} = i \hbar \frac{dA^u_{nm}(\vec{k})}{dt} \rho_{nn}(\vec{k}, t) + i \hbar \frac{dA_{nm}^u(\vec{k})}{dt} \rho_{mm}(\vec{k}, t) = [a^*_n(\vec{k}), H] a_m(\vec{k}) + [a_n(\vec{k}), H] a^*_m(\vec{k})
\] (11)

For the intrinsic NLO effect from geometry of eigenstates, we ignore all the electron-electron, electron-phonon, and electron-impurity scattering term here. We expand the density matrix up to the second order of field strength,

\[
\rho_{nm}(\vec{k}, t) = \rho_{nm}^{(0)}(\vec{k}) + \rho_{nm}^{(1)}(\vec{k}, t) + \rho_{nm}^{(2)}(\vec{k}, t) + \ldots,
\] (12)

in which \( \rho_{nm}(\vec{k}, t) \propto |E|^\lambda \), and \( \rho_{nm}^{(0)}(\vec{k}) = \delta_{nm} \rho_{nm}^{(0)}(\vec{k}) \) is the electronic distribution of ground state. Here, \( \rho_{nn}^{(0)} = \frac{1}{1 + \exp(-\frac{\epsilon_n}{k_B T})} (k_B T) \text{Boltzmann constant, } T \text{ temperature} \) is fermi-Dirac distribution of band \( n \). From Eq. (11), the first-order frequency dependent inter-band (\( n \neq m \)) density matrix reads,

\[
\rho_{nm}^{(1)}(\vec{k}, \omega) = \frac{e E(\omega) \cdot \tilde{A}_{nm}(\vec{k}) [\rho_{nm}^{(0)}(\vec{k}) - \rho_{nn}^{(0)}(\vec{k})]}{\hbar \omega + \epsilon_n(\vec{k}) + i \eta}
\] (13)

where \( \eta \) is an infinitesimal parameter. The first-order intra-band frequency dependent density matrix reads

\[
\rho_{nn}^{(1)}(\vec{k}, \omega) = -\frac{ie}{\hbar \omega} E(\omega) \cdot \partial_\omega \rho_{nn}^{(0)}(\vec{k}).
\] (14)

From Eq. (11), the second-order frequency dependent intra-band and inter-band density matrices read,
\[
\rho^{(2)}_{nm}(\omega_3) = \frac{-ie}{\hbar \omega_3} \sum_{\omega_1, \omega_2} \delta(\omega_3, \omega_1 + \omega_2) \tilde{E}(\omega_1) \cdot \{ \partial_k \rho^{(1)}_{nm}(\omega_2) + i \sum_m [\tilde{A}_{mn}(\tilde{k})\rho^{(1)}_{mn}(\tilde{k}, \omega_2) - \rho^{(1)}_{nm}(\omega_2)\tilde{A}_{nm}(\tilde{k})] \},
\]
\[
\rho^{(2)}_{nm}(\tilde{k}, \omega_3) = \frac{-ie}{\hbar \omega_3 + \epsilon_{nm}} \sum_{\omega_1, \omega_2} \delta(\omega_3, \omega_1 + \omega_2) \tilde{E}(\omega_1) \cdot \{ \partial_k \rho^{(1)}_{nm}(\tilde{k}, \omega_2) + i \sum_l [\tilde{A}_{ln}(\tilde{k})\rho^{(1)}_{lm}(\tilde{k}, \omega_2) - \rho^{(1)}_{nl}(\tilde{k}, \omega_2)\tilde{A}_{ml}(\tilde{k})] \},
\]

respectively.

**DC current** Up to second order response, the interband dc current reads,
\[
J^{\text{inter}}_u(0) = \frac{e^3}{\hbar} \sum_{\alpha, \beta} \sum_{\omega} E_\alpha(-\omega)E_\beta(\omega)
\sum_{n,m} \int d\tilde{k} \partial_\alpha G^\alpha_{mn}(\tilde{k}) A^\alpha_{nm}(\tilde{k}) (\rho^{(0)}_{mn}(\tilde{k}) - \rho^{(0)}_{nm}(\tilde{k})).
\]

In the case of resonant excitation, \( \hbar \omega = \epsilon_{nm} \), the interband dc current reads,
\[
J^{\text{inter}}_u(0) = \frac{i \pi e^3}{\hbar^2} \sum_{\alpha, \beta} \sum_{\omega} E_\alpha(-\omega)E_\beta(\omega)
\sum_{n,m} \int d\tilde{k} \partial_\alpha A^\alpha_{mn}(\tilde{k}) A^\beta_{nm}(\tilde{k}) (\rho^{(0)}_{mn}(\tilde{k}) - \rho^{(0)}_{nm}(\tilde{k})).
\]

For linearly polarized light, \( E_\alpha(-\omega)E_\beta(\omega) = E_\alpha(\omega)E_\beta(-\omega) = \frac{1}{2} \vert E(\omega) \vert^2 \), the second order optical conductivity reads,
\[
\sigma^{(2)}_{\alpha\beta}(0) = \frac{\pi e^3}{\hbar^2} \sum_{\alpha, \beta} \sum_m \int d\tilde{k} \partial_\alpha G^\beta_{mn}(\tilde{k}) \rho^{(0)}_{mn}(\tilde{k}),
\]
in which \( F^{\alpha\beta}_{mn}(\tilde{k}) = \partial_\alpha A^\alpha_{mn}(\tilde{k}) + \partial_\beta A^\alpha_{mn}(\tilde{k}) \) is the antisymmetric Berry curvature in momentum space. Eq. (19) demonstrates that the CPGE is determined by the integral of gradient of Berry curvature in momentum space. In the system with spatial inversion symmetry and TRIS, \( F^{\alpha\beta}_{mn}(\tilde{k}) \) is vanishing \([1]\), and \( A^\alpha_{mn}(\tilde{k}) \) is constant. Therefore, both LPGE and CPGE are vanishing if the system has both TRIS and spatial inversion symmetry.

Eqs. (18) and (19) are main discoveries of this work, and they demonstrate that LPGE and CPGE have underlying connection with the symmetric and antisymmetric parts of QGT in momentum space, respectively. The nonlinear Hall effect \([30]\), i.e., the direction of dc current is perpendicular to the plane of electric field, can exist under both linearly and circularly polarized field. Especially, under circularly polarized light, \( \sigma^{(2)}_{\alpha\beta}(0) \) can be approximately quantized in chiral topological semimetal, if only the electron around the \( \Gamma \) point is excited by light with low frequency. For example, in the band structure of chiral topological semimetals RbSi and CoSi\([13, 51]\), the Berry curvature accumulated around the \( \Gamma \) point is \( F^{xy}_{mn}(\tilde{k}) = \lambda k_z/|k|^3 \), and \( \partial_z F^{xy}_{mn}(\tilde{k}) = \lambda/|k|^3 \), where \( \lambda \) is the Chern number of targeted band. Therefore, second order conductivity \( \sigma^{(2)}_{xy}(0) \) is proportional to \( \lambda \), and affords a simple approach to detect the Chern number of targeted band if circularly polarized with appropriate frequency is applied.

In summary, we explored the underlying connection between GPE and QGT. We concluded that the gradient of the symmetric part of QGT is related to the LPGE, while the gradient of antisymmetric part (Berry curvature) is related to the CPGE. Our work afforded an alternative interpretation for PGE in the view of QGT, and classified the underlying connection between CPGE (LPGE) and Berry curvature (quantum metric). The CPGE can be approximately quantized in chiral topological semimetals under circularly polarized with appropriate frequency.

ZL is supported by the National Natural Science Foundation of China (11604068). TI and TT are supported by MEXT via Exploratory Challenge on Post-K Computer (Frontiers of Basic Science: Challenging the Limits). The calculations were performed on the Hokusai system (Project No. Q20246) of Riken. H.B.S. is grateful for support from the Society of Interdisciplinary Research (SOIRREE) and HKUST (IGN17SC04; R9418).
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