特邀综述

Implication of Berry Phase in Nonlinear Transport Phenomena

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Electron transport involves a family of important material properties that can provide a wealth of information about the underlying system. In recent few decades, it has been more and more realized that the geometrical quantities in the momentum space, such as the Berry curvature and the orbital magnetic moment, play a highly involving role in the electron transport. Their connection to the transport property has been examined to great extent at the linear order. At the nonlinear order, such connection has begun to be examined recently. This not only deepens the understanding of such nonlinear transport phenomena, but also helps the band-structure engineering of them. In this work, we introduce the general semiclassical framework of the transport theory, which can be used to study both the linear and nonlinear phenomena, and exemplify the Berry phase effect in several transport phenomena.

贝利相与非线性输运

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电子输运现象包含一系列很重要的材料性质，并可以用来提供关于载体物理系统的很多信息。在最近的二三十年，人们逐渐认识到除了电子能谱之外，电子在布里渊区的几何性质，比如贝利曲率和轨道磁矩，会在电子输运中起到关键性的作用。在线性输运现象中的此种关联已引起广泛的兴趣并得到深入的研究。然而，在非线性输运现象中的电子几何性质的作用的研究最近才逐渐起步。此种关联可以大大加深对各种非线性输运现象的认识，并对如何从材料控制上调节非线性现象的强度提供有价值的研究视角和指导原则。基于此背景，本文试图引入研究输运现象的半经典理论框架，并在几种输运现象中举例说明贝利相的作用。

I. INTRODUCTION

After its introduction by M. Berry [1], the concept of Berry phase is found to have profound implications in physics. Especially, the electron transport theory greatly benefits from a deeper understanding of such geometric quantities in the momentum space induced by the wave functions instead of the energy spectrum. One of the most well-known examples is the anomalous Hall effect [2, 3]. Central to the anomlaous Hall effect is the concept of the Berry curvature, i.e. the Berry phase per unit area in the momentum space. Its integration yields the Chern number, which can be used for the topological classification of matter [4]. Moreover, it can also be used to study the optical transport phenomena, such as the circular dichroism in ferromagnetic materials [5], the valley-contrast circular dichroism [6], and so on. The Berry curvature is also the analog of the magnetic field in the momentum space. As such, the source of Berry curvature is the momentum-space analog of magnetic monopoles, which can be realized by Weyl/Dirac points, leading to unique magnetoresistance behavior [7, 8].

Recently, it has been realized that besides linear transport phenomena, the geometrical quantities are also essential in understanding various nonlinear transport phenomena. For example, in noncentrosymmetric materials, the second harmonic generation and nonlinear anomalous Hall effect is mainly determined by the dipole of the Berry curvature in the momen-

tum space [9–12]. Weyl semimetals can have a quantized circular photogalvanic current due to the quantized Berry curvature dipole [13]. Berry phase and Berry connection helps the discovery of the large photovoltaic effect in Weyl semimetals [14–16]. Two spatially dispersive phenomena, i.e. the natural optical activity and the directional dichroism, depend on the dipole of magnetic moment and quantum metric tensor [17–20], respectively.

In this work, we introduce several efforts of understanding the transport phenomena in the language of Berry phase and Berry curvature in a systematic framework, i.e. the semiclassical theory, paying special attention to the nonlinear phenomena. Our work is organized as follows. In Sect. II, we provide a self-contained introduction of the Berry phase and Berry curvature and exemplify them using a simple two-band model. In Sect. III, we briefly introduce the semiclassical theory up to linear order of electromagnetic fields and hence discuss the role of Berry phase and Berry curvature in characterizing the order parameter as well as the static and optical anomalous Hall effect. In Sect. IV, we extend the semiclassical theory up to second order and exemplify the implication of the Berry phase and Berry curvature in nonlinear phenomena in static and optical transport.

II. GENERAL SETTING OF BERRY PHASE AND CURVATURE

In this section, we provide a self-contained introduction to the Berry phase and Berry curvature. We will start from the demonstration of Berry phase in the adiabatic evolution during which the driven parameter returns to its starting value after some time τ. This Berry phase can be re-expressed using the integration of the Berry curvature through the Stoke's theorem. The Berry curvature is then exemplified in a two-band model which is of great importance to later application.

A. Berry phase

We consider a physical system with the Hamiltonian $\hat{H}_0(\boldsymbol{R})$ subject to the evolution of the parameter $\boldsymbol{R}(t)$. We assume that after a period of time τ, $\boldsymbol{R}$ returns to the initial value: $\boldsymbol{R}(\tau) = \boldsymbol{R}(0)$. Moreover, $\boldsymbol{R}(t)$ varies slowly enough to warrant the adiabatic approximation.

In this setting, M. V. Berry discussed the following famous question [1]. If initially the system resides in an eigenstate $|n_0\rangle$ of $\hat{H}_0(\boldsymbol{R}(0))$, the adiabaticity of the evolution guarantees that approximately at any subsequent time t, the system is in the instantaneous eigenstate $|n(\boldsymbol{R})\rangle$, determined by the following equation

$$\hat{H}_0(\boldsymbol{R})|n(\boldsymbol{R})\rangle = \varepsilon_n(\boldsymbol{R})|n(\boldsymbol{R})\rangle. \tag{1}$$

Then at time $t = \tau$ when $\boldsymbol{R}$ returns to its starting value, $|n(\boldsymbol{R})\rangle$ returns to $|n_0\rangle$. However, to what extent does the final state conform to $|n_0\rangle$?

To proceed, one first makes the following observation: Eq. (1) only determines the eigenstate up to a phase factor. Without loss of generality, one can assume that the intermediate state during the evolution has the following form:

$$|\Psi\rangle = e^{i\gamma}|n(\boldsymbol{R})\rangle. \tag{2}$$

On the other hand, $|\Psi\rangle$ should satisfy the Schrödinger equation

$$i\frac{d}{dt}|\Psi\rangle = \hat{H}_0(\boldsymbol{R})|\Psi\rangle. \tag{3}$$

This yields the equation for the phase factor

$$\frac{d\gamma}{dt} = \dot{\boldsymbol{R}}(t) \cdot \langle n(\boldsymbol{R})|i\boldsymbol{\nabla}_{\boldsymbol{R}}|n(\boldsymbol{R})\rangle - \varepsilon_n(\boldsymbol{R}). \tag{4}$$

One can define the phase factor apart from the well-known dynamical factor

$$\frac{d\tilde{\gamma}}{dt} = \frac{d\gamma}{dt} + \varepsilon_n(\boldsymbol{R}) = \dot{\boldsymbol{R}}(t) \cdot \langle n(\boldsymbol{R})|i\boldsymbol{\nabla}_{\boldsymbol{R}}|n(\boldsymbol{R})\rangle. \tag{5}$$

The accumulated phase is then

$$\begin{aligned}
\tilde{\gamma} &= \int_0^\tau \dot{\boldsymbol{R}}(t) \cdot \langle n(\boldsymbol{R})|i\boldsymbol{\nabla}_{\boldsymbol{R}}|n(\boldsymbol{R})\rangle dt \\
&= \oint \langle n(\boldsymbol{R})|i\boldsymbol{\nabla}_{\boldsymbol{R}}|n(\boldsymbol{R})\rangle \cdot d\boldsymbol{R}.
\end{aligned} \tag{6}$$

The integrant in the above equation is defined as the Berry connection

$$\mathcal{A}(\boldsymbol{R}) = \langle n(\boldsymbol{R})|i\boldsymbol{\nabla}_{\boldsymbol{R}}|n(\boldsymbol{R})\rangle. \tag{7}$$

It is easy to check that the Berry connection is not gauge-independent. Instead, adding an arbitrary phase factor $e^{-i\phi}$ ($U(1)$ gauge transformation) to $|n(\boldsymbol{R})\rangle$ will also shift the Berry connection:

$$\mathcal{A}(\boldsymbol{R}) \to \mathcal{A}(\boldsymbol{R}) + \boldsymbol{\nabla}_{\boldsymbol{R}}\phi. \tag{8}$$

Interestingly, the resulting Berry phase is gauge-independent modulo 2π in a cyclic evolution. Since the wave function is single-valued, the phase is subject to the following constraint

$$\phi[\boldsymbol{R}(\tau)] - \phi[\boldsymbol{R}(0)] = 2\pi m, \tag{9}$$

with m being an arbitrary integer. After the gauge transformation, the change in Berry phase reads

$$\tilde{\gamma} \to \tilde{\gamma} + \phi[\boldsymbol{R}(\tau)] - \phi[\boldsymbol{R}(0)] = \tilde{\gamma} + 2m\pi. \tag{10}$$

Therefore, the phase factor $e^{i\tilde{\gamma}}$ is unchanged.

Three comments are in order about the derivation of the Berry phase. First, we note that Eq. (6) does not depend on the changing rate of $\boldsymbol{R}(t)$. It only depends on the geometry of the path in the parameter space. From this regard, the Berry phase is also referred to as the geometric phase.

Second, the adiabaticity in the derivation is to guarantee the cyclic evolution of states arising from the cyclic changing of parameters. However, the appearance of the Berry phase implies that this cyclic evolution of states only exists in the projective Hilbert space, in which eigenstates with different but constant phase factors are identified as the same point.

Third, the real wave function $|\Psi\rangle$ as defined in Eq. (2) is parallel transported during the evolution. This can be seen by multiplying $\langle\Psi|$ to Eq. (3)

$$\langle\Psi|i\frac{d}{dt}|\Psi\rangle = \langle\Psi|\hat{H}_0(\boldsymbol{R})|\Psi\rangle = 0. \tag{11}$$

The second equality can be realized by either shifting the energy level to 0 or by discounting the dynamical phase factor.

This implies that at each infinitesimal and consecutive step of evolution, $|\Psi(\boldsymbol{R})\rangle$ and $|\Psi(\boldsymbol{R} + \Delta\boldsymbol{R})\rangle$ are in phase. Yet the final state is deviated from the initial state by a path-dependent Berry phase. This is actually an array of consecutive and infinitesimal realization of the Pancharatnam's phase [21, 22], which states that if both states $|1\rangle$ and $|2\rangle$ are in phase with another state $|3\rangle$, i.e. $|1\rangle + |3\rangle$ and $|2\rangle + |3\rangle$ have the maximum magnitude [23], $|1\rangle$ and $|2\rangle$ are not necessarily in phase, because they can have different components that are orthogonal to $|3\rangle$.

Based on the above understandings, the concept of the Berry phase has been generalized in several scenarios. First, the adiabaticity can be removed if the cyclic evolution in the projective Hilbert space is already ensured, such as the periodic spin procession in a magnetic field. This leads to the Ahanorov-Anandan phase [24], which discusses the geometric phase accumulated in a cyclic, but not necessarily adiabatic, evolution. This generalization can be most easily seen through the parallel transport in Eq. (11). Assume that $|\Psi(t = \tau)\rangle = e^{i\phi}|\Psi(t = 0)\rangle$. One can thus define $|\tilde{\Psi}\rangle = e^{-if(t)}|\Psi\rangle$, with $f(\tau) - f(0) = \phi$ and hence $|\tilde{\Psi}(t = \tau)\rangle = |\tilde{\Psi}(t = 0)\rangle$. Thus $|\tilde{\Psi}\rangle$ is a well-defined single-valued wave function which plays the role of $|n(\boldsymbol{R})\rangle$ in the original setting. The Berry phase is then expressed as

$$\phi = \int_0^\tau dt \langle \tilde{\Psi} | i \frac{d}{dt} | \tilde{\Psi} \rangle. \tag{12}$$

We comment that enlightened by the Pancharatnam's phase, the cyclic condition can be further removed [25]: a non-cyclic evolution can be made cyclic by adding an auxiliary evolution along the geodesic curve.

Second, the Berry phase is also extended to the non-Abelian case [26]. Suppose that initially the system is in the eigenstate $|n_1\rangle$, which is one in a set of degenerate states $|n_i\rangle$, $i = 1, 2, \cdots$. Adiabaticity ensures that the intermediate states have the following form

$$|\Psi\rangle = U_{i1}(t)|n_i(t)\rangle. \tag{13}$$

Here and hereafter, repeated indices are summed. Using the parallel transport law in Eq. (11), one can then obtain the equation for the unitary evolution opeartor

$$U_{i1} = i\mathcal{A}_{ij} U_{j1}, \tag{14}$$

where $\mathcal{A}_{ij} = \langle n_i | i(d/dt) | n_j \rangle$ is the nonabelian Berry connection. Letting the initial state run through the whole degenerate set of states, we obtain

$$U = i\mathcal{A}U \Rightarrow U = Pe^{i\int_0^\tau dt\mathcal{A}}, \tag{15}$$

where P stands for path-ordering. In case of nondegenerate single-band scenario, the above result naturally returns to the Abelian Berry phase.

B. Berry curvature

Although the Berry connection $\mathcal{A}$ is gauge-dependent, one can define a gauge-independent quantity called Berry curvature as follows

$$\Omega_{ij} = \partial_{R_i} \mathcal{A}_j - \partial_{R_j} \mathcal{A}_i. \tag{16}$$

This is similar to the relation between the magnetic field and the gauge-dependent vector potential in the electromagnetic theory. Indeed, under the gauge transformation, the Berry connection transforms according to Eq. (8), indicating that

$$\Omega_{ij} \to \partial_{R_i}(\mathcal{A}_j + \partial_{R_j}\phi) - \partial_{R_j}(\mathcal{A}_i + \partial_{R_i}\phi) = \Omega_{ij}. \tag{17}$$

If the parameter space is three-dimensional, since the Berry curvature is antisymmetric about the two indices, it can be represented by a axial vector

$$\boldsymbol{\Omega} = \frac{1}{2}\epsilon_{ijk}\Omega_{ij}\hat{e}_k. \tag{18}$$

In terms of the Berry curvature, the Berry phase can be recast using the Stokes' theorem

$$\tilde{\gamma} = \oint_\Sigma \mathcal{A} \cdot d\boldsymbol{R} = \int_S \boldsymbol{\Omega} \cdot d\boldsymbol{S}. \tag{19}$$

Here S is a surface enclosed by the closed curve Σ. This makes the gauge-independence of the Berry phase more evident.

The Berry curvature is an essential ingredient in our later demonstration. For calculation convenience, we will put it in an alternate form.

$$\begin{aligned}
\Omega_{ij}^n &= \partial_i\langle n|i\partial_j|n\rangle - (i \leftrightarrow j) \\
&= i\langle\partial_i n|\partial_j n\rangle - (i \leftrightarrow j) \\
&= \sum_m i\langle\partial_i n|m\rangle\langle m|\partial_j n\rangle - (i \leftrightarrow j) \\
&= -\sum_{m\neq n} i\langle n|\partial_i m\rangle\langle m|\partial_j n\rangle - (i \leftrightarrow j) \\
&= i\sum_{m\neq n} \frac{\langle n|\partial_i \hat{H}_0|m\rangle\langle m|\partial_j \hat{H}_0|n\rangle - (i \leftrightarrow j)}{(\varepsilon_n - \varepsilon_m)^2}.
\end{aligned} \tag{20}$$

In the above derivation, we use the identity

$$\begin{aligned}
\langle n|\partial_i \hat{H}_0|m\rangle &= -\varepsilon_m\langle\partial_i n|m\rangle - \varepsilon_n\langle n|\partial_i m\rangle \\
&= (\varepsilon_m - \varepsilon_n)\langle n|\partial_i|m\rangle.
\end{aligned} \tag{21}$$

Here ∂_i is short for ∂_{R_i} and we also recover the level index n for the Berry curvature.

Equation (20) demonstrates the importance of the residue Hilbert space $|m\rangle$ with $m \neq n$. This is in accordance with the interpretation of the Berry phase as a consecutive and infinitesimal array of the Pancharatnam's phase, whose existence lies in the different orthogonal parts. If there is no residue Hilbert space, i.e. we sum the Berry curvature over different levels, we recover the conservation law for the Berry curvature

$$\sum_n \Omega_{ij}^n = 0. \tag{22}$$

Finally, we comment that the Berry curvature is part of a more general geometrical quantity called quantum geometrical tensor [27], defined as follows

$$G_{ij}^n = \langle\partial_i n|\partial_j n\rangle - \mathcal{A}_i^n\mathcal{A}_j^n. \tag{23}$$

The imaginary part of G_{ij}^n yields the Berry curvature while its real part yields the quantum metric tensor g_{ij}^n: $G_{ij}^n = g_{ij}^n - $

$i\Omega_{ij}^n/2$. The quantum metric tensor can also be expressed using summation of states:

$$
\begin{aligned}
g_{ij}^n &= -\mathrm{Re} \sum_{m \neq n} \langle n|\partial_i m\rangle\langle m|\partial_j n\rangle \\
&= \mathrm{Re} \sum_{m \neq n} \frac{\langle n|\partial_i \hat{H}_0|m\rangle\langle m|\partial_j \hat{H}_0|n\rangle}{(\varepsilon_n - \varepsilon_m)^2} .
\end{aligned} \tag{24}
$$

The meaning of the quantum metric tensor can be interpreted as follows: we consider two neighbouring Bloch states $|n(\boldsymbol{R})\rangle$ and $|n(\boldsymbol{R} + \delta\boldsymbol{R})\rangle$ and define their distance as follows

$$
\begin{aligned}
d ={}& \langle n(\boldsymbol{R} + \delta\boldsymbol{R}) - n(\boldsymbol{R})|n(\boldsymbol{R} + \delta\boldsymbol{R}) - n(\boldsymbol{R})\rangle \\
& - |\langle n(\boldsymbol{R})|n(\boldsymbol{R} + \delta\boldsymbol{R}) - n(\boldsymbol{R})\rangle|^2 .
\end{aligned} \tag{25}
$$

The second term is to remove the gauge-dependence. This distance can be expressed using the quantum metric tensor:

$$
d = g_{ij}^n dR_i dR_j . \tag{26}
$$

Therefore, the quantum metric tensor measures the distance of neighbouring Bloch states in the Hilbert space. The Berry curvature and quantum metric tensor generally modify the dynamical structure and correct the energy in the evolution, respectively, as shown in the study of the lattice motion under the Born-Oppenheimer approximation.

C. Generic two-level system

As a concrete example, we consider a generic two-level system. Despite its simple structure, it has wide applicability as it can be realized in various different scenarios as discussed later. The model Hamiltonian is given by

$$
\hat{H} = h_x\sigma_x + h_y\sigma_y + h_z\sigma_z , \tag{27}
$$

where h_x, h_y, and h_z are real parameters and $\boldsymbol{\sigma}$ is the Pauli matrix for spin/pseudospin. Its eigenenergy reads $\varepsilon = \pm h$ with $h = \sqrt{h_x^2 + h_y^2 + h_z^3}$. The corresponding wave function is

$$
\psi_+ = \frac{1}{\xi}\begin{pmatrix} h_x - ih_y \\ h - h_z \end{pmatrix} , \psi_- = \frac{1}{\xi}\begin{pmatrix} h_z - h \\ h_x + ih_y \end{pmatrix} , \tag{28}
$$

where $\xi = \sqrt{2h(h - h_z)}$ is a normalization factor.

We consider the Berry curvature in the vector form for the lower band in the parameter space spanned by $\boldsymbol{h} = (h_x, h_y, h_z)$. Without loss of generality, we calculate the z-component, which can be expressed as follows

$$
\begin{aligned}
\Omega_z &= i\frac{\langle\psi_-|\partial_x \hat{H}_0|\psi_+\rangle\langle\psi_+|\partial_y \hat{H}_0|\psi_-\rangle - (x \leftrightarrow y)}{(\varepsilon_- - \varepsilon_+)^2} \\
&= -\frac{1}{2h^2}\mathrm{Im}[\langle\psi_-|\sigma_x|\psi_+\rangle\langle\psi_+|\sigma_y|\psi_-\rangle] .
\end{aligned} \tag{29}
$$

The interband elements for the pseudospin can be obtained from the wave function and the results read

$$
\langle\psi_-|\sigma_x|\psi_+\rangle = \frac{h_x^2 - h_y^2 - (h - h_z)^2 - 2ih_xh_y}{2h(h - h_z)}
$$

$$
\langle\psi_-|\sigma_y|\psi_+\rangle = \frac{2h_xh_y + ih_x^2 - ih_y^2 + i(h - h_z)^2}{2h(h - h_z)} . \tag{30}
$$

Then the Berry curvature can be calculated and the result is

$$
\Omega_z = \frac{h_z}{2h^3} \Rightarrow \boldsymbol{\Omega} = \frac{\boldsymbol{h}}{2h^3} . \tag{31}
$$

This is the field generated by the monopole at $h = 0$. Indeed, one can integrate the Berry curvature over a spherical surface that enclose the origin to find

$$
\frac{1}{2\pi}\int_S d\boldsymbol{S} \cdot \boldsymbol{\Omega} = 1 . \tag{32}
$$

This resembles the Gauss' theorem that connects the field strength with the source charge.

III. TRANSPORT PHENOMENA AT LINEAR ORDER

In this section, we discuss various transport phenomena related the concept of Berry phase and Berry curvature. We will focus on the semiclassical framework as introduced in Sect. III A, because it is intuitive and has clear physical interpretation. In Sect. III B, we discuss how to obtain the material order parameter up to linear order, i.e. electric polarization and magnetization. The understanding of the polarization also benefits from a more general formulation of the semiclassical theory, which yields the charge pumping and charge density modulation, as shown in Sect. III C. In Sect. III D, We discuss one of the most important applications of the Berry curvature, i.e. the anomalous Hall effect. We then explore two phenomena closely related to the anomalous Hall effect. We first change the driven force of currents from electric field to the thermal gradient and derive the anomalous thermoelectric current in Sect. III E. Finally, we consider the counterpart of the anomalous Hall effect in optics, i.e. the circular dichroism in ferromagnetic materials, in Sect. III F.

A. Semiclassical framework of electron dynamics in electromagnetic fields

We consider the following general crystal Hamiltonian in the non-relativistic limit that governs the dynamics of Bloch electrons under uniform electromagnetic fields

$$
\hat{H}_{\mathrm{f}} = \hat{H}_0[\hat{\boldsymbol{p}} + e\boldsymbol{A}(\boldsymbol{r}); \boldsymbol{r}] + e\boldsymbol{E} \cdot \boldsymbol{r} , \tag{33}
$$

where $\hat{H}_0(\hat{\boldsymbol{p}}; \boldsymbol{r})$ is the unperturbed Hamiltonian for periodic crystals with $\hat{\boldsymbol{p}}$ being the momentum operator. We emphasize that the magnetic vector potential $\boldsymbol{A}$ is different from the Berry connection $\mathcal{A}$. Here the magnetic field modifies the Hamiltonian through the minimal coupling and we ignore the Zeeman coupling for simplicity. In the case of magnetic field only, generally speaking, $\hat{H}_{\mathrm{f}}$ only respects the translational symmetry when the magnetic flux through a unit cell is a rational number times the flux quantum [28–31].

The dynamics of Bloch electrons can in principle be solved from $\hat{H}_{\mathrm{f}}$. But for simplicity, we assume that the solution to the Schrödinger equation $(i\hbar\partial_t - \hat{H}_{\mathrm{f}})\psi = 0$ has the form of a wave packet. To deal with the unbounded position operator, in constructing the wave packet, one can decompose the full Hamiltonian into a local one plus perturbations. The local Hamiltonian $\hat{H}_c$ is obtained by evaluating the electromagnetic

potential in $\hat{H}_f$ at the center of mass position $\boldsymbol{r}_c$ of the wave packet. As a result, $\hat{H}_c$ recovers the translational symmetry of the original unperturbed crystal. The resulting local Hamiltonian reads:

$$\hat{H}_c = \hat{H}_0[\hat{\boldsymbol{p}} + e\boldsymbol{A}(\boldsymbol{r}_c); \boldsymbol{r}] + e\boldsymbol{E} \cdot \boldsymbol{r}_c, \tag{34}$$

Its eigenenergy is the local Bloch bands $\varepsilon_n[\boldsymbol{p} + (e/\hbar)\boldsymbol{A}(\boldsymbol{r}_c)] + e\boldsymbol{E} \cdot \boldsymbol{r}_c$, where n is the band index and $\hbar\boldsymbol{p}$ the crystal momentum. Its eigenfunction is the Bloch function $e^{i\boldsymbol{p}\cdot\boldsymbol{r}}|u_n[\boldsymbol{p} + e\boldsymbol{A}(\boldsymbol{r}_c)/\hbar]\rangle$.

In the following, we focus on a single band with index 0 and assume that it is well separated from all the other bands. Under weak electromagnetic fields, a Bloch electron starting from some state in one band will stay in the same band. Therefore, the wave packet is constructed as the superposition of Bloch states from the band 0 [3, 32]

$$|W\rangle = \int d\boldsymbol{p}\, C_0(\boldsymbol{p}) e^{i\boldsymbol{p}\cdot\boldsymbol{r}}|u_0[\boldsymbol{p} + e\boldsymbol{A}(\boldsymbol{r}_c)/\hbar]\rangle. \tag{35}$$

$|W\rangle$ should be normalized, indicating $\int d\boldsymbol{p}|C_0|^2 = 1$.

The coefficient C_0 is constrained in two ways. On one hand, we assume that the wave packet is sharply localized in the momentum space. Therefore, the magnitude of C_0 satisfies $|C_0|^2 \approx \delta(\boldsymbol{p} - \boldsymbol{p}_c)$. Here $\hbar\boldsymbol{p}_c$ is the center of mass momentum of the wave packet, i.e.

$$\boldsymbol{p}_c = \langle W|\boldsymbol{p}|W\rangle. \tag{36}$$

On the other hand, the center of mass position $\boldsymbol{r}_c$ of the wave packet should be determined in a self-consistent manner in the following way [32]:

$$\boldsymbol{r}_c = \langle W|\boldsymbol{r}|W\rangle$$
$$= \int d\boldsymbol{p}d\boldsymbol{p}'\, C_0^\star(\boldsymbol{p}) C_0(\boldsymbol{p}')\langle u_0(\boldsymbol{p})|(i\partial_{\boldsymbol{p}}e^{-i\boldsymbol{p}\cdot\boldsymbol{r}})e^{i\boldsymbol{p}'\cdot\boldsymbol{r}}|u_0(\boldsymbol{p}')\rangle$$

$$= \left.\frac{\partial\gamma}{\partial\boldsymbol{p}}\right|_{\boldsymbol{p}=\boldsymbol{p}_c} + \mathcal{A}_0(\boldsymbol{k}_c), \tag{37}$$

where $\gamma = -arg(C_0)$, $\mathcal{A}_0(\boldsymbol{p}) = \langle u_0(\boldsymbol{p})|i\partial_{\boldsymbol{p}}|u_0(\boldsymbol{p})\rangle$ is the intraband Berry connection, and $\hbar\boldsymbol{k}_c = \hbar\boldsymbol{p}_c + e\boldsymbol{A}(\boldsymbol{r}_c)$ is the gauge-independent physical momentum. We have ignored the dependence on $\boldsymbol{A}(\boldsymbol{r}_c)$ in $|u_0(\boldsymbol{p})\rangle$ for simple notation. These two constraints complete the construction of the wave packet. We comment that the construction of the wave packet can also be generalized to account for the general multiband case [33, 34].

The dynamics of the wave packet can be derived using the variational principle. The coefficient C_0 in the wave packet can be determined through the variation of the Lagrangian evaluated under the wave packet state. Alternatively, C_0 has independent magnitude and phase, which are determined by $\boldsymbol{k}_c$ (or equivalently, $\boldsymbol{p}_c$) and $\boldsymbol{r}_c$ respectively, and hence one can also obtain the dynamics of $\boldsymbol{r}_c$ and $\boldsymbol{k}_c$, i.e. the phase space equations of motion, from the variation of the Lagrangian.

The Lagrangian reads

$$L = \langle W|i\hbar\partial_t - \hat{H}_c - \hat{H}_1|W\rangle. \tag{38}$$

where $\hat{H}_1 = \frac{e}{2}\boldsymbol{B} \cdot [(\boldsymbol{r} - \boldsymbol{r}_c) \times \hat{\boldsymbol{v}}] + e\boldsymbol{E} \cdot (\boldsymbol{r} - \boldsymbol{r}_c)$ is the first order correction to the local Hamiltonian $\hat{H}_c$ with $\hat{\boldsymbol{v}}$ being the velocity operator. Here we have used the symmetric gauge for the magnetic vector potential. The energy part can be easily evaluated

$$\langle W|\hat{H}_c|W\rangle = \int d\boldsymbol{p}d\boldsymbol{p}'\, C_0(\boldsymbol{p})^\star C_0(\boldsymbol{p}')\langle u_0(\boldsymbol{p})|e^{-i\boldsymbol{p}\cdot\boldsymbol{r}}\hat{H}_c e^{i\boldsymbol{p}'\cdot\boldsymbol{r}}|u_0(\boldsymbol{p}')\rangle$$
$$= \int d\boldsymbol{p}d\boldsymbol{p}'\, C_0(\boldsymbol{p})^\star C_0(\boldsymbol{p}')\varepsilon_0(\boldsymbol{p})\delta(\boldsymbol{p} - \boldsymbol{p}')$$
$$= \int d\boldsymbol{p}\,\delta(\boldsymbol{p} - \boldsymbol{p}_c)\varepsilon_0(\boldsymbol{p})$$
$$= \varepsilon_0(\boldsymbol{k}_c). \tag{39}$$

The other part of correction to energy is

$$\langle W|\hat{H}_1|W\rangle = \langle W|\frac{e}{2}\boldsymbol{B} \cdot [(\boldsymbol{r} - \boldsymbol{r}_c) \times \hat{\boldsymbol{v}}] + e\boldsymbol{E} \cdot (\boldsymbol{r} - \boldsymbol{r}_c)|W\rangle$$
$$= \frac{e}{2}\boldsymbol{B} \cdot \langle W|(\boldsymbol{r} - \boldsymbol{r}_c) \times \hat{\boldsymbol{v}}|W\rangle$$
$$= \frac{e}{2}\boldsymbol{B} \cdot \int d\boldsymbol{p}d\boldsymbol{p}'\, C_0(\boldsymbol{p})^\star C_0(\boldsymbol{p}')\langle u_0(\boldsymbol{p})|e^{-i\boldsymbol{p}\cdot\boldsymbol{r}}(\boldsymbol{r} - \boldsymbol{r}_c) \times \hat{\boldsymbol{v}}e^{i\boldsymbol{p}'\cdot\boldsymbol{r}}|u_0(\boldsymbol{p}')\rangle$$
$$= \frac{e}{2}\boldsymbol{B} \cdot \int d\boldsymbol{p}d\boldsymbol{p}'\, C_0(\boldsymbol{p})^\star C_0(\boldsymbol{p}')\langle u_0(\boldsymbol{p})|(i\partial_{\boldsymbol{p}}e^{-i\boldsymbol{p}\cdot\boldsymbol{r}}) \times \hat{\boldsymbol{v}}e^{i\boldsymbol{p}'\cdot\boldsymbol{r}}|u_0(\boldsymbol{p}')\rangle - \frac{e}{2}\boldsymbol{B} \cdot \boldsymbol{r}_c \times \boldsymbol{v}_0$$
$$= \frac{e}{2}\boldsymbol{B} \cdot \int d\boldsymbol{p}(-i\partial_{\boldsymbol{p}})C_0(\boldsymbol{p})^\star C_0(\boldsymbol{p}) \times \boldsymbol{v}_0 + \frac{e}{2}\boldsymbol{B} \cdot \int d\boldsymbol{p} C_0(\boldsymbol{p})^\star C_0(\boldsymbol{p})(-i)\langle\partial_{\boldsymbol{p}}u_0(\boldsymbol{p})| \times \hat{\boldsymbol{v}}|u_0(\boldsymbol{p})\rangle - \frac{e}{2}\boldsymbol{B} \cdot \boldsymbol{r}_c \times \boldsymbol{v}_0. \tag{40}$$

Here the first term and the intraband part in the second term exactly cancel the third term, using the definition of the center of mass position $\boldsymbol{r}_c$ in Eq. (37). The remaining term reads

$$\langle W|\hat{H}_1|W\rangle$$
$$= \frac{e}{2}\boldsymbol{B} \cdot \sum_{n\neq 0} \int d\boldsymbol{p}|C_0(\boldsymbol{p})|^2(-i)\langle\partial_{\boldsymbol{p}}u_0|u_n\rangle \times \langle u_n|\hat{\boldsymbol{v}}|u_0\rangle$$

$$= \frac{e}{2}\boldsymbol{B} \cdot \mathrm{Re}\sum_{n\neq 0} \mathcal{A}_{0n} \times \boldsymbol{v}_{n0}, \tag{41}$$

where $\mathcal{A}_{0n} = \langle u_0|i\partial_{\boldsymbol{p}}|u_n\rangle$ is the interband Berry connection and $\boldsymbol{v}_{n0} = \langle u_n|\hat{\boldsymbol{v}}|u_0\rangle$ is the interband velocity matrix element. If we define $\boldsymbol{m}_0 = -\frac{e}{2}\mathrm{Re}\sum_{n\neq 0}\mathcal{A}_{0n} \times \boldsymbol{v}_{n0}$ as the orbital magnetic moment, this energy correction takes the form of the Zeeman coupling due to the orbital motion.

It is interesting to find that in a two-band model, the orbital

magnetic moment is connected to the Berry curvature. In fact, we have

$$
\begin{aligned}
\boldsymbol{m}_0 &= -\frac{e}{2}\mathrm{Re}\mathcal{A}_{0n}\times\boldsymbol{v}_{n0}\\
&= \frac{e}{2}(\varepsilon_0-\varepsilon_n)\mathrm{Re}[i\mathcal{A}_{0n}\times\mathcal{A}_{n0}]\\
&= \frac{e}{2}(\varepsilon_0-\varepsilon_n)\boldsymbol{\Omega}_0\,.
\end{aligned}
\tag{42}
$$

This indicates that the magnetic moment is proportional to the Berry curvature with a factor linear in the band gap. The conservation law of the Berry curvature implies that for a two-band model, the upper and lower band have opposite Berry curvatures. Due to the appearance of the band gap, they have exactly the same orbital magnetic moment. As a result, under a magnetic field, they will shift in the same direction in energy.

One interesting fact is that if one begins with the relativistic Dirac Hamiltonian where the magnetic field only enters through the minimal coupling and constructs a coherent wave packet for the upper two bands, one can naturally obtain the spin magnetic moment as the orbital magnetic moment of this wave packet [35]. Mathematically, this is equivalent to the Foldy-Wouthuysen transformation that reduces the Dirac Hamiltonian to the non-relativistic Schrödinger Hamiltonian [36, 37]. Here we have a heuristic and clear picture for the emergence of spin using the semiclassical theory.

We now calculate the time derivative in the Lagrangian:

$$
\begin{aligned}
\langle W|i\hbar\partial_t|W\rangle &= \int d\boldsymbol{p}d\boldsymbol{p}'\, C_0(\boldsymbol{p})^{\star}C_0(\boldsymbol{p}')\langle u_0(\boldsymbol{p})|e^{-i\boldsymbol{p}\cdot\boldsymbol{r}}i\hbar d_t e^{i\boldsymbol{p}'\cdot\boldsymbol{r}}|u_0(\boldsymbol{p}')\rangle\\
&\quad + \int d\boldsymbol{p}d\boldsymbol{p}'\, C_0(\boldsymbol{p})^{\star}[i\hbar d_t C_0(\boldsymbol{p}')]\langle u_0(\boldsymbol{p})|e^{-i\boldsymbol{p}\cdot\boldsymbol{r}}e^{i\boldsymbol{p}'\cdot\boldsymbol{r}}|u_0(\boldsymbol{p}')\rangle\\
&= [d_t\boldsymbol{A}(\boldsymbol{r}_c)]\cdot\int d\boldsymbol{p}\, C_0(\boldsymbol{p})^{\star}C_0(\boldsymbol{p})\langle u_0(\boldsymbol{p})|i\partial_{\boldsymbol{p}}|u_0(\boldsymbol{p}')\rangle + \int d\boldsymbol{p}d\boldsymbol{p}'\, C_0(\boldsymbol{p})^{\star}[\hbar d_t\gamma(\boldsymbol{p},\boldsymbol{r}_c,t)]C_0(\boldsymbol{p})\\
&= \frac{e}{2}\boldsymbol{B}\times\dot{\boldsymbol{r}}_c\cdot\mathcal{A}_0 + [\hbar d_t\gamma(\boldsymbol{p},\boldsymbol{r}_c,t)]|_{\boldsymbol{p}=\boldsymbol{p}_c}\\
&= \frac{e}{2}\boldsymbol{B}\times\dot{\boldsymbol{r}}_c\cdot\mathcal{A}_0 + \hbar d_t\gamma(\boldsymbol{p}_c,\boldsymbol{r}_c,t) - \hbar\dot{\boldsymbol{p}}_c\cdot\boldsymbol{\partial}_{\boldsymbol{p}_c}\gamma(\boldsymbol{p}_c,\boldsymbol{r}_c,t)\\
&= \hbar d_t\gamma(\boldsymbol{p}_c,\boldsymbol{r}_c,t) - \hbar\dot{\boldsymbol{p}}_c\cdot(\boldsymbol{r}_c-\mathcal{A}_0) + \frac{e}{2}\boldsymbol{B}\times\dot{\boldsymbol{r}}_c\cdot\mathcal{A}_0\\
&= d_t\gamma(\boldsymbol{p}_c,\boldsymbol{r}_c,t) - \hbar\dot{\boldsymbol{k}}_c\cdot(\boldsymbol{r}_c-\mathcal{A}_0) + \frac{e}{2}\boldsymbol{B}\times\dot{\boldsymbol{r}}_c\cdot\boldsymbol{r}_c\,.
\end{aligned}
\tag{43}
$$

In the above derivation, we use the fact that the phase of C_0 is generally a function of $\boldsymbol{p}$, $\boldsymbol{r}_c$, and t (although its magnitude depends on $\boldsymbol{p}_c$). The total time derivative does not affect the dynamics and will be ignored. We can sum up the above contributions and obtain the following total Langrangian

$$
L = -(\boldsymbol{r}_c-\mathcal{A}_0)\cdot\hbar\dot{\boldsymbol{k}}_c - \frac{1}{2}e\boldsymbol{B}\times\boldsymbol{r}_c\cdot\dot{\boldsymbol{r}}_c - \tilde{\varepsilon}_0\,,
\tag{44}
$$

where $\tilde{\varepsilon} = \varepsilon_0 - \boldsymbol{B}\cdot\boldsymbol{m}_0 + e\boldsymbol{E}\cdot\boldsymbol{r}_c$ is the modified band energy up to first order.

The Berry connection $\mathcal{A}_0$ is an essential ingredient in the Lagrangian. In the absence of $\mathcal{A}_0$, the above Lagrangian reduces to the conventional canonical form for electrons under electromagnetic fields:

$$
L = \left(\hbar\boldsymbol{k}_c - \frac{1}{2}e\boldsymbol{B}\times\boldsymbol{r}_c\right)\cdot\dot{\boldsymbol{r}}_c - \tilde{\varepsilon}_0\,.
\tag{45}
$$

The appearance of $\mathcal{A}_0$ indicates that $\boldsymbol{r}_c$ and $\boldsymbol{k}_c$ are not canonical variables. To make this statement clearer, we consider the following transformation [35]

$$
\boldsymbol{r}_c = \boldsymbol{q} + \mathcal{A}_0 + \frac{1}{2}\frac{e}{\hbar}(\boldsymbol{B}\times\mathcal{A}_0\cdot\boldsymbol{\partial}_{\boldsymbol{p}})\mathcal{A}_0 + \frac{1}{2}\frac{e}{\hbar}\boldsymbol{\Omega}_0\times(\boldsymbol{B}\times\mathcal{A}_0)\,,
\tag{46}
$$

$$
\boldsymbol{k}_c = \boldsymbol{p} + \frac{1}{2}\frac{e}{\hbar}\boldsymbol{B}\times\boldsymbol{q} + \frac{e}{\hbar}\boldsymbol{B}\times(\boldsymbol{r}_c-\boldsymbol{q})\,,
\tag{47}
$$

where $\boldsymbol{\Omega}_0 = \boldsymbol{\nabla}_{\boldsymbol{p}}\times\mathcal{A}_0$ is the Berry curvature. Here the argument of $\mathcal{A}_0$ and $\boldsymbol{\Omega}_0$ is $\boldsymbol{p} + \frac{e}{2\hbar}\boldsymbol{B}\times\boldsymbol{q}$. Then the Lagrangian in Eq. (44) recovers the canonical form for $\boldsymbol{p}$ and $\boldsymbol{q}$

$$
L = \hbar\boldsymbol{p}\cdot\dot{\boldsymbol{q}} - \tilde{\varepsilon}_0\,.
\tag{48}
$$

Eq. (46) and (47) thus describe the connection between physical variables and canonical variables.

One consequence of the noncanonicality is that the phase space measure for the volume element $d\boldsymbol{r}_c d\boldsymbol{k}_c$ has to change based on the Jacobian $\mathcal{J} = \partial(\boldsymbol{q},\boldsymbol{p})/\partial(\boldsymbol{r}_c,\boldsymbol{k}_c)$, i.e. the phase space density of states is $\mathcal{D} = |\det\mathcal{J}|$. To calculate this, we notice that the Lie bracket can be expressed using the above matrices

$$
\{\xi_i,\xi_j\} = \mathcal{J}^{-1}\begin{pmatrix}0 & I\\ -I & 0\end{pmatrix}(\mathcal{J}^{-1})^T\,,
\tag{49}
$$

with $\boldsymbol{\xi} = (\boldsymbol{r}_c,\boldsymbol{k}_c)$. As a result

$$
\det\{\xi_i,\xi_j\} = (\det\mathcal{J})^{-2}\,.
\tag{50}
$$

On the other hand, we have

$$
\{\xi_i,\xi_j\} = \begin{pmatrix}R & Q\\ S & T\end{pmatrix}\,,
\tag{51}
$$

with

$$R = \begin{pmatrix} 0 & \Omega_z & -\Omega_y \\ -\Omega_z & 0 & \Omega_x \\ \Omega_y & -\Omega_x & 0 \end{pmatrix} \tag{52}$$

$$Q = \begin{pmatrix} 1 - \frac{e}{\hbar}(B_z\Omega_z + B_y\Omega_y) & 0 & 0 \\ 0 & 1 - \frac{e}{\hbar}(B_z\Omega_z + B_x\Omega_x) & 0 \\ 0 & 0 & 1 - \frac{e}{\hbar}(B_x\Omega_x + B_y\Omega_y) \end{pmatrix} \tag{53}$$

$$S = \begin{pmatrix} -1 + \frac{e}{2\hbar}(B_z\Omega_z + B_y\Omega_y) & 0 & 0 \\ 0 & -1 + \frac{e}{2\hbar}(B_z\Omega_z + B_x\Omega_x) & 0 \\ 0 & 0 & -1 + \frac{e}{2\hbar}(B_x\Omega_x + B_y\Omega_y) \end{pmatrix} \tag{54}$$

$$T = \begin{pmatrix} 0 & -eB_z/\hbar & eB_y/\hbar \\ eB_z/\hbar & 0 & -eB_x/\hbar \\ -eB_y/\hbar & eB_x/\hbar & 0 \end{pmatrix}. \tag{55}$$

This yields the following phase space density of states [38]

$$\mathcal{D} = 1 + \frac{e}{\hbar} \boldsymbol{B} \cdot \boldsymbol{\Omega}_0(\boldsymbol{k}_c). \tag{56}$$

Since the range for the physical position and momentum are the crystal volume and the Brillouin zone, respectively, it is natural to evaluate the statistical average of any operator in the physical phase space spanned by $(\boldsymbol{r}_c, \boldsymbol{k}_c)$. Therefore, we should always use the modified density of states $\mathcal{D}$.

The noncanonicality of $\boldsymbol{r}_c$ and $\boldsymbol{k}_c$ as shown in Eq. (51) will affect the structure of the equations of motion. From Eq. (44), the Euler-Lagrangian equations of motion yield the following phase space dynamics [32]

$$\frac{\partial L}{\partial \boldsymbol{k}_c} = \frac{d}{dt}\frac{\partial L}{\partial \dot{\boldsymbol{k}}_c} \Rightarrow \dot{\boldsymbol{r}}_c = \frac{\partial \tilde{\varepsilon}_0}{\hbar \partial \boldsymbol{k}_c} - \dot{\boldsymbol{k}}_c \times \boldsymbol{\Omega}_0(\boldsymbol{k}_c), \tag{57}$$

$$\frac{\partial L}{\partial \boldsymbol{r}_c} = \frac{d}{dt}\frac{\partial L}{\partial \dot{\boldsymbol{r}}_c} \Rightarrow \hbar\dot{\boldsymbol{k}}_c = -e\boldsymbol{E} - e\dot{\boldsymbol{r}}_c \times \boldsymbol{B}. \tag{58}$$

The second term in the velocity equation is the anomalous velocity due to the noncanonicality. It has the same form with the Lorentz force from the magnetic field. From this perspective, the Berry curvature is also the momentum-space analog of the magnetic field. The above equations of motion have wide applicability in the various transport phenomena.

B. Electromagnetic Dipoles

The semiclassical theory can be used to derive the electromagnetic multipoles in crystals. We note that because the wave packet is sharply localized in the momentum space, it should have a finite width in the real space. The charge and current distributions within this spread can be inhomogeneous. To describe such internal inhomogeneity of the wave packet, it is natural to use electromagnetic multipoles. It is interesting that such multipoles are related to the macroscopic order parameters of the bulk materials, such as the electric polarization and magnetization, eventually.

The electric dipole of the wave packet reads $\langle W| - e\boldsymbol{r}|W\rangle = -e\boldsymbol{r}_c$, with $\boldsymbol{r}_c$ given in Eq. (37). Under the periodic gauge [3, 39], the Bloch wave function and the coefficient C_0 is periodic in the momentum space. Therefore, if we integrate $\boldsymbol{r}_c$ in the Brillouin zone, the first term in Eq. (37) from the phase of C_0

can at most yield an integer multiples of 2π and hence can be ignored. The remaining term yields the electric polarization in crystals

$$\boldsymbol{P} = -e \int \frac{d\boldsymbol{k}_c}{(2\pi)^3} \boldsymbol{\mathcal{A}}_0(\boldsymbol{k}_c). \tag{59}$$

The electric polarization can also be derived using either the adiabatic charge pumping current [40, 41] or the dipole correction to the charge density [3]. Here our method yields the same electric polarization under the periodic gauge for Bloch functions.

The magnetic dipole has already been derived in Eq. (40). Here we further demonstrate that it directly contributes to the orbital magnetization. We consider the free energy under magnetic field. On one hand, the band energy is modified through the Zeeman coupling between the orbital magnetic moment and the magnetic field. On the other hand, magnetic field also changes the phase space density of states. They together yield the following free energy

$$F = \int \mathcal{D}\frac{d\boldsymbol{k}}{(2\pi)^3}(-k_BT)\ln\left[1 + \exp\left(\frac{\tilde{\varepsilon}_0 - \mu}{k_BT}\right)\right]. \tag{60}$$

Be definition, the orbital magnetization reads [42]

$$\boldsymbol{M} = -\left.\frac{\partial F}{\partial \boldsymbol{B}}\right|_{B=0} = \int \frac{d\boldsymbol{k}}{(2\pi)^3}\left(f_0\boldsymbol{m}_0 - \frac{e}{\hbar}g_e\boldsymbol{\Omega}_0\right), \tag{61}$$

where $g_e = -(k_BT)\ln[1+\exp((\varepsilon_0-\mu)/k_BT)]$ is the grand potential density, and f_0 is the equilibrium Fermi distribution function. Here and hereafter we will ignore the subscript c in the integration over $\boldsymbol{k}_c$. This is the contribution of the orbital magnetization from band 0. The total contribution can be obtained by summing over the band index. In the two contributions to the orbital magnetization, the first one is due to the relative motion inside the wave packet, or the self-rotation of the wave packet; the second one is due to the rotation of the wave packet as a whole, or the revolution of the wave packet. The same magnetization can also be obtained through the Wannier function approach [43, 44], the exact Hofstadter spectrum [45, 46], or the linear response theory [47, 48].

C. General perturbation

In this section, we derive the electron dynamics under general perturbations. We consider the following Hamiltonian

$$\hat{H}_{\mathrm{f}} = \hat{H}(\hat{\pmb{p}}, \pmb{r}; \pmb{r}, t)\,. \tag{62}$$

The first and second argument $\pmb{r}$ represent fast and slowly changing part of the Hamiltonian, respectively. We also assume that the system evolves slowly with time. We can expand the slowly changing part of the Hamiltonian around the center of the wave packet.

$$\hat{H}_{\mathrm{f}} = \hat{H}_{\mathrm{c}} + \hat{H}_1 + \cdots$$
$$\hat{H}_{\mathrm{c}} = \hat{H}(\hat{\pmb{p}}, \pmb{r}; \pmb{r}_c, t)\,,$$
$$\hat{H}_1 = \frac{1}{2}\partial_{\pmb{r}_c}\hat{H}_{\mathrm{c}} \cdot (\pmb{r} - \pmb{r}_c) + h.c.. \tag{63}$$

The eigenenergy and eigenstate of the local Hamiltonian generally have the following form: $\varepsilon_n(\pmb{p}, \pmb{r}_c, t)$ and $e^{i\pmb{p}\cdot\pmb{r}}|u_n(\pmb{p}, \pmb{r}_c, t)\rangle$.

The derivation of the Lagrangian is very similar to that under electromagnetic fields. For the energetic part, it is easy to check that

$$\langle W|\hat{H}_{\mathrm{c}}|W\rangle = \varepsilon_0(\pmb{p}_c, \pmb{r}_c, t)\,, \tag{64}$$
$$\langle W|\hat{H}_1|W\rangle = \sum_{n\neq 0}\mathrm{Re}[\langle u_0|\partial_{\pmb{r}_c}\hat{H}_{\mathrm{c}}|u_n\rangle\langle u_n|i\partial_{\pmb{p}_c}|u_0\rangle]\,. \tag{65}$$

We can recover Eq. (41) from the second equation by noting that $\partial_{\pmb{r}_c}\hat{H}_c = -(e/2)\pmb{B}\times\hat{\pmb{v}}$ under magnetic field. For the dynamic part, one can find

$$\langle W|i\hbar\partial_t|W\rangle = \dot{\pmb{r}}_c \cdot \langle u_0|i\partial_{\pmb{r}_c}|u_0\rangle + \langle u_0|i\partial_t|u_0\rangle + \hbar d_t\gamma(\pmb{p}_c, \pmb{r}_c, t) - \hbar\dot{\pmb{p}}_c \cdot \partial_{\pmb{p}_c}\gamma(\pmb{p}_c, \pmb{r}_c, t) \tag{66}$$

Therefore, the Lagrangian reads

$$L = \dot{\pmb{r}}_c \cdot \mathcal{A}^{\pmb{r}} + \mathcal{A}^t - \hbar\dot{\pmb{p}}_c \cdot (\pmb{r}_c - \mathcal{A}^{\pmb{p}}) - \tilde{\varepsilon}\,, \tag{67}$$

where we have define $\mathcal{A}^{\pmb{r}} = \langle u_0|i\partial_{\pmb{r}_c}|u_0\rangle$, $\mathcal{A}^t = \langle u_0|i\partial_t|u_0\rangle$, $\mathcal{A}^{\pmb{p}} = \langle u_0|i\partial_{\pmb{p}_c}|u_0\rangle$, and $\tilde{\varepsilon} = \varepsilon_0(\pmb{p}_c, \pmb{r}_c, t) + \sum_{n\neq 0}\mathrm{Re}[\langle u_0|\partial_{\pmb{r}_c}\hat{H}_{\mathrm{c}}|u_n\rangle\langle u_n|i\partial_{\pmb{p}_c}|u_0\rangle]$. The dynamics can be obtained through the Euler-Lagrangian equations of motion

$$\frac{\partial L}{\partial \pmb{p}_c} = \frac{d}{dt}\frac{\partial L}{\partial \dot{\pmb{p}}_c}$$
$$\Rightarrow \dot{\pmb{r}}_c = \frac{\partial \tilde{\varepsilon}_0}{\hbar\partial \pmb{p}_c} - (\pmb{\Omega}^{\pmb{p}\pmb{r}} \cdot \dot{\pmb{r}}_c + \pmb{\Omega}^{\pmb{p}\pmb{p}} \cdot \dot{\pmb{p}}_c) - \pmb{\Omega}^{\pmb{p}t}\,, \tag{68}$$
$$\frac{\partial L}{\partial \pmb{r}_c} = \frac{d}{dt}\frac{\partial L}{\partial \dot{\pmb{r}}_c}$$
$$\Rightarrow \hbar\dot{\pmb{p}}_c = -\frac{\partial \tilde{\varepsilon}}{\partial \pmb{r}_c} + (\pmb{\Omega}^{\pmb{r}\pmb{r}} \cdot \dot{\pmb{r}}_c + \pmb{\Omega}^{\pmb{r}\pmb{p}} \cdot \dot{\pmb{p}}_c) + \pmb{\Omega}^{\pmb{r}t}\,, \tag{69}$$

where $\pmb{\Omega}^{\pmb{r}\pmb{r}}_{ij} = \partial_{\pmb{r}_{ci}}\mathcal{A}^{\pmb{r}}_j - \partial_{\pmb{r}_{cj}}\mathcal{A}^{\pmb{r}}_i$ is the real space Berry curvature, $\pmb{\Omega}^{\pmb{p}\pmb{p}}_{ij} = \partial_{\pmb{p}_{ci}}\mathcal{A}^{\pmb{p}}_j - \partial_{\pmb{p}_{cj}}\mathcal{A}^{\pmb{p}}_i$ is the momentum space Berry curvature, $\pmb{\Omega}^{\pmb{p}\pmb{r}}_{ij} = \partial_{\pmb{p}_{ci}}\mathcal{A}^{\pmb{r}}_j - \partial_{\pmb{r}_{cj}}\mathcal{A}^{\pmb{p}}_i$ is the mixed Berry curvature, $\pmb{\Omega}^{\pmb{r}\pmb{p}}_{ij} = -\pmb{\Omega}^{\pmb{p}\pmb{r}}_{ij}$, $\pmb{\Omega}^{\pmb{p}t} = \partial_{\pmb{p}_c}\mathcal{A}^t - \partial_t\mathcal{A}^{\pmb{p}}$, and $\pmb{\Omega}^{\pmb{r}t} = \partial_{\pmb{r}_c}\mathcal{A}^t - \partial_t\mathcal{A}^{\pmb{r}}$.

One can make several interesting observations. First, in the case of electromagnetic field, the $\pmb{r}_c$-dependence in the energy is simply the electrostatic potential and $\pmb{\Omega}^{\pmb{r}\pmb{r}}$ will reduce to the magnetic field (i.e. the magnetic field can be viewed as the real

space Berry curvature). Then the above equations of motion will coincide with those in Eq. (57) and (58).

Second, one can obtain the electric polarization from different perspectives. On one hand, we can consider a system slowly changing in time but homogeneous in space. Then we can obtain the current in the system by integrating the electron velocity

$$\pmb{J} = e\int \frac{d\pmb{k}}{8\pi^3}\pmb{\Omega}^{\pmb{p}t}\,. \tag{70}$$

This is the charge pumping current. To gain further insight, we note that the periodic part of the Bloch function can be added to an arbitrary phase factor. Specifically, one can choose a gauge so that $\mathcal{A}^t$ is periodic in the Brillouin zone and its derivative with respect to $\pmb{p}$ will integrate to zero. Therefore, the current can be put in the following form

$$\pmb{J} = -e\partial_t\int \frac{d\pmb{k}}{8\pi^3}\mathcal{A}^{\pmb{p}}\,. \tag{71}$$

Together with the displacement current in the Maxwell equations, which suggests $\partial_t\pmb{P} \to \pmb{J}$, we can identify

$$\pmb{P} = -e\int \frac{d\pmb{k}}{8\pi^3}\mathcal{A}^{\pmb{p}}\,. \tag{72}$$

This is the same as the polarization obtained in the last section.

On the other hand, we can consider a spatially inhomogeneous but static system. As a result, $\pmb{\Omega}^{\pmb{r}\pmb{p}}$ and $\pmb{\Omega}^{\pmb{p}\pmb{r}}$ are nonzero. The Lie bracket in Eq. (51) becomes (here we only keep entries up to first order spatial derivative)

$$\{\xi_i, \xi_j\} = \begin{pmatrix} 0 & \Omega_{xy}^{pp} & -\Omega_{zx}^{pp} & 1-\Omega_{xx}^{pr} & 0 & 0 \\ -\Omega_{xy}^{pp} & 0 & \Omega_{yz}^{pp} & 0 & 1-\Omega_{yy}^{pr} & 0 \\ \Omega_{zx}^{pp} & -\Omega_{yz}^{pp} & 0 & 0 & 0 & 1-\Omega_{zz}^{pr} \\ -1+\Omega_{xx}^{pr} & 0 & 0 & 0 & 0 & 0 \\ 0 & -1+\Omega_{yy}^{pr} & 0 & 0 & 0 & 0 \\ 0 & 0 & -1+\Omega_{zz}^{pr} & 0 & 0 & 0 \end{pmatrix}. \tag{73}$$

Therefore, the density of states read

$$\mathcal{D} = 1 + \mathrm{Tr}\boldsymbol{\Omega}^{pr}. \tag{74}$$

With this density of states, we can calculate the electron density

$$\begin{aligned} \rho &= -e \int \frac{d\boldsymbol{k}}{(2\pi)^3}(1 + \mathrm{Tr}\boldsymbol{\Omega}^{pr}) \\ &= \rho_0 + e\boldsymbol{\partial}_{\boldsymbol{r}} \cdot \int \frac{d\boldsymbol{k}}{(2\pi)^3}\mathcal{A}^p. \end{aligned} \tag{75}$$

The second equality is true when we choose the gauge such that $\mathcal{A}^r$ is periodic in the Brillouin zone. The contribution from the polarization to the electron density reads $-\boldsymbol{\nabla} \cdot \boldsymbol{P}$. Therefore, we can identify the polarization as

$$\boldsymbol{P} = -e \int \frac{d\boldsymbol{k}}{(2\pi)^3}\mathcal{A}^p. \tag{76}$$

D. Anomalous Hall effect

An important application of the semiclassical theory is to understand the anomalous Hall effect, which refers to a Hall-type current in ferromagnets solely driven by an electric field and without the presence of the magnetic field. It is a topic under extensive studies. For a review, see Ref. [2]. In the past, there are three mechanisms identified: the intrinsic contribution, the skew-scattering contribution, and the side-jump contribution. Here we focus on the intrinsic contribution. The anomalous velocity in Eq. (57) contributes to a current density

$$\boldsymbol{J} = -\frac{e^2}{\hbar} \int \frac{d\boldsymbol{k}}{(2\pi)^3}\boldsymbol{E} \times \boldsymbol{\Omega}. \tag{77}$$

For $\boldsymbol{\Omega}$ along $\hat{z}$ direction, we have the following anomalous Hall conductivity

$$\sigma_{xy} = -\sigma_{yx} = -\frac{e^2}{\hbar} \int \frac{d\boldsymbol{k}}{(2\pi)^3}\Omega_z. \tag{78}$$

It is interesting to note that the conventional Hall conductivity is due to the magnetic field, i.e. the magnetic field bends the trajectory of Bloch electrons, while the anomalous Hall conductivity is due to the Berry curvature, which is the momentum-space analog of the magnetic field and which generates a velocity transverse to the driven force. Moreover, we comment that the anomalous Hall conductivity is intrinsic, i.e. independent of the relaxation process, while the conventional Hall conductivity is quadratic in the transport relaxation time.

To further understand the symmetry requirement of the anomalous Hall conductivity, we analyze the transformation of the Berry curvature under symmetry operations. This can be most easily seen from Eq. (57). Both $\dot{\boldsymbol{r}}_c$ and $\dot{\boldsymbol{k}}_c$ flip sign under inversion operation. Therefore, the inversion symmetry requires $\boldsymbol{\Omega}(\boldsymbol{k}) = \boldsymbol{\Omega}(-\boldsymbol{k})$. $\dot{\boldsymbol{r}}_c$ and $\dot{\boldsymbol{k}}_c$ transform oppositely under time reversal operation, indicating $\boldsymbol{\Omega}(-\boldsymbol{k}) = -\boldsymbol{\Omega}(\boldsymbol{k})$ [49] under time reversal symmetry. This indicates that anomalous Hall effect can only happen when the time reversal symmetry is broken, while the conventional Hall effect does not require the system to break any symmetry.

As a concrete example, we consider the following low-energy Hamiltonian.

$$\hat{H} = \lambda(k_x\sigma_y - k_y\sigma_x) + \Delta\sigma_z, \tag{79}$$

where $\boldsymbol{\sigma}$ is the spin Pauli matrix, k_x and k_y are momentum along x and y direction. The first term is a Rashba-type spin-orbit coupling. The second term is the Zeeman coupling due to the local net spin moment. The energy spectrum is $\varepsilon_\pm = \pm\sqrt{\lambda^2 k^2 + \Delta^2}$. The Zeeman coupling breaks the time-reversal symmetry. However, we also need the spin-orbit coupling to have the anomalous Hall current as it admits the orbital motion sense the broken time-reversal symmetry in the spin space. Using Eq. (20), we can calculate the Berry curvature, which reads

$$\Omega_\pm = \pm\frac{\lambda^2\Delta}{2(\lambda^2 k^2 + \Delta^2)^{3/2}}. \tag{80}$$

When the Fermi level μ falls in the valence band, one can integrate the Berry curvature and find the anomalous Hall conductivity

$$\sigma_{xy} = -\frac{e^2}{2h}\frac{\Delta}{|\mu|}. \tag{81}$$

We note that both ferromagnets and antiferromangets breaks the time-reversal symmetry. For a long time, only ferromagnetic metals with net magnetization are under study for the anomalous Hall effect. Recently, it is also shown that antiferromagnetic metals can also generate the anomalous Hall effect [50–58]. This can also be explained by the symmetry property of the Berry curvature. Without loss of generality, we still consider the xy-component of the conductivity. We have shown that its existence requires the time reversal symmetry to be broken. Moreover, we note that for Ω_z, $\dot{\boldsymbol{r}}_c$ and $\dot{\boldsymbol{k}}_c$ in the velocity equation transform oppositely under mirror-x or mirror-y symmetry, indicating that Ω_z is odd under mirror-x or mirror-y operations. Therefore, σ_{xy} also needs those symmetries to be broken. A net magnetization along $\hat{z}$ direction would certainly do that, but it is not necessary. As shown in Ref. [56], there are antiferromagnets that also satisfy this criteria and hence make the anomalous Hall current possible.

Finally, we comment that the derivation of the anomalous Hall current using semiclassical theory also helps the understanding of other types of Hall current. In writing Eq. (77), we use the fact that the current density in the phase space has the form $-e\dot{r}_c$. If we change the electric charge to other types of charge, we can obtain different types of current due to the anomalous velocity. For example, in the gapped graphene system, one can use the valley index τ instead of the electric charge, which will gives rise to the valley Hall current, indicating that electrons in different valleys will move oppositely and hence accumulate at opposite edges [59, 60]. One can also use one of the spin component instead of the electric charge, giving rise to the spin Hall current [33, 61, 62], which suggests accumulation of opposite spins at opposite edges.

E.   Using wave packet for spatial average

Central to the electromagnetic theory is the macroscopic Maxwell equations, which can be derived from the microscopic Maxwell equations by applying a proper spatial average technique. We note that this is for localized charge and current sources [63]. In solid state physics, we often deal with unbounded crystals. In this regard, we can use the wave packet as the smallest unit in the macroscopic level to perform the spatial average.

The spatial average process proceeds as follows. We consider an arbitrary operator $\hat{O}$ and multiply it with a sampling

function (delta function) to obtain the density of the operator

$$O(\boldsymbol{q}) = \int \frac{d\boldsymbol{r}_c d\boldsymbol{k}_c}{(2\pi)^3} f(\boldsymbol{r}_c, \boldsymbol{k}_c) \langle W|\hat{O}\delta(\boldsymbol{r}-\boldsymbol{q})|W\rangle. \tag{82}$$

In writing this density, we use the distribution of the wave packet to substitute the real distribution function. Each wave packet has a localized position $\boldsymbol{r}_c$ and we can expand the sampling function around it:

$$\begin{aligned}
\delta(\boldsymbol{r}-\boldsymbol{q}) &= \delta(\boldsymbol{r}-\boldsymbol{r}_c+\boldsymbol{r}_c-\boldsymbol{q}) \\
&= \delta(\boldsymbol{r}_c-\boldsymbol{q}) + (\boldsymbol{r}_c-\boldsymbol{r})\cdot\partial_{\boldsymbol{q}}\delta(\boldsymbol{q}-\boldsymbol{r}_c) + \cdots. \tag{83}
\end{aligned}$$

Plugging this expansion into the density of the operator, we obtain

$$\begin{aligned}
O(\boldsymbol{q}) &= \int \frac{d\boldsymbol{r}_c d\boldsymbol{k}_c}{(2\pi)^3} f(\boldsymbol{r}_c, \boldsymbol{k}_c)\langle W|\hat{O}|W\rangle\delta(\boldsymbol{q}-\boldsymbol{r}_c) \\
&\quad + \partial_{\boldsymbol{q}}\cdot\int \frac{d\boldsymbol{r}_c d\boldsymbol{k}_c}{(2\pi)^3} f(\boldsymbol{r}_c, \boldsymbol{k}_c)\langle W|\hat{O}(\boldsymbol{r}_c-\boldsymbol{r})|W\rangle\delta(\boldsymbol{q}-\boldsymbol{r}_c) \\
&= \int \frac{d\boldsymbol{k}_c}{(2\pi)^3} f(\boldsymbol{q}, \boldsymbol{k}_c)\langle u_0|\hat{O}|u_0\rangle \\
&\quad - \partial_{\boldsymbol{q}}\cdot\int \frac{d\boldsymbol{k}_c}{(2\pi)^3} f(\boldsymbol{q}, \boldsymbol{k}_c)\boldsymbol{D}(\boldsymbol{O}), \tag{84}
\end{aligned}$$

where $\boldsymbol{D}(\boldsymbol{O})$ is the dipole moment of the operator $\hat{O}$. In deriving the above result, we use the fact that $\langle W|\hat{O}|W\rangle = \langle u_0|\hat{O}|u_0\rangle$ [64] and

$$\begin{aligned}
\langle W|\hat{O}(\boldsymbol{r}-\boldsymbol{r}_c)|W\rangle &= \int d\boldsymbol{p}d\boldsymbol{p}' C_0(\boldsymbol{p})^\star C_0(\boldsymbol{p}')\langle u_0(\boldsymbol{p})|e^{-i\boldsymbol{p}\cdot\boldsymbol{r}}\hat{O}(\boldsymbol{r}-\boldsymbol{r}_c)e^{i\boldsymbol{p}'\cdot\boldsymbol{r}}|u_0(\boldsymbol{p}')\rangle \\
&= \int d\boldsymbol{p}d\boldsymbol{p}' C_0(\boldsymbol{p})^\star C_0(\boldsymbol{p}')\langle u_0(\boldsymbol{p})|e^{-i\boldsymbol{p}\cdot\boldsymbol{r}}\hat{O}(-i\partial_{\boldsymbol{p}'})e^{i\boldsymbol{p}'\cdot\boldsymbol{r}}|u_0(\boldsymbol{p}')\rangle - \boldsymbol{r}_c\langle u_0|\hat{O}|u_0\rangle \\
&= \int d\boldsymbol{p} C_0(\boldsymbol{p})^\star(i\partial_{\boldsymbol{p}})C_0(\boldsymbol{p})\langle u_0|\hat{O}|u_0\rangle + \int d\boldsymbol{p} C_0(\boldsymbol{p})^\star C_0(\boldsymbol{p})i\langle u_0|\hat{O}|\partial_{\boldsymbol{p}}u_0\rangle - \boldsymbol{r}_c\langle u_0|\hat{O}|u_0\rangle \\
&= \mathrm{Re}\sum_{n\neq 0}\langle u_0|\hat{O}|u_n\rangle\langle u_n|i\partial_{\boldsymbol{p}}|u_0\rangle\Big|_{\boldsymbol{p}=\boldsymbol{k}_c}. \tag{85}
\end{aligned}$$

This completes the spatial average process.

The result in Eq. (84) has been used in the study of the charge and spin transport [42, 65]. Here we will focus on the charge transport. For this purpose, we put $\hat{O} = -e\hat{v}_i$, i.e. the local current operator. Then we have

$$\begin{aligned}
\boldsymbol{D}(\boldsymbol{O}) &= -e\mathrm{Re}\sum_{n\neq 0}\langle u_0|\hat{v}_i|u_n\rangle(\mathcal{A}_j)_{n0}\hat{e}_j \\
&= -\frac{e}{\hbar}\mathrm{Re}\sum_{n\neq 0} i(\varepsilon_0-\varepsilon_n)(\mathcal{A}_i)_{0n}(\mathcal{A}_j)_{n0}\hat{e}_j \\
&= -\frac{e}{2\hbar}\mathrm{Re}\sum_{n\neq 0} i(\varepsilon_0-\varepsilon_n)[(\mathcal{A}_i)_{0n}(\mathcal{A}_j)_{n0} - (i\leftrightarrow j)]\hat{e}_j \\
&= -\epsilon_{ijk}m_k\hat{e}_j. \tag{86}
\end{aligned}$$

Here we have used the identity in Eq. (21) and $\boldsymbol{m}$ is the orbital magnetic moment as defined in Eq. (41). Therefore, the local

current density reads

$$\boldsymbol{J} = -e\int \frac{d\boldsymbol{k}}{(2\pi)^3} f(\boldsymbol{r}, \boldsymbol{k})\boldsymbol{v}_0 + \boldsymbol{\nabla}\times\int \frac{d\boldsymbol{k}}{(2\pi)^3} f(\boldsymbol{r}, \boldsymbol{k})\boldsymbol{m}. \tag{87}$$

On the other hand, if the orbital magnetization $\boldsymbol{M}$ is nonzero, due to the inhomogeneous distribution function f, $\boldsymbol{M}$ will also vary in space, which can give rise to a magnetization current from $\boldsymbol{\nabla}\times\boldsymbol{M}$. This magnetization current does not contribute to transport and has to be discounted from the total current [42, 66]. The resulting transport current is

$$\begin{aligned}
\boldsymbol{J}_{\mathrm{tr}} &= \boldsymbol{J} - \boldsymbol{\nabla}\times\boldsymbol{M} \\
&= \frac{e}{\hbar}\boldsymbol{\nabla}\times\int \frac{d\boldsymbol{k}}{(2\pi)^3} g_e\boldsymbol{\Omega}_0. \tag{88}
\end{aligned}$$

Here we have use the orbital magnetization given in Eq. (61).

In the presence of a thermal gradient, we have

$$\boldsymbol{\nabla}g_e = \frac{\boldsymbol{\nabla}T}{T}[g_e - (\varepsilon_0-\mu)f]. \tag{89}$$

Therefore, we have the following thermoelectric current

$$J_{\text{tr}} = \frac{e}{\hbar} \frac{\nabla T}{T} \times \int \frac{dk}{(2\pi)^3} [g_e - (\varepsilon_0 - \mu)f]\Omega_0 . \qquad (90)$$

This current always flows normal to the thermal gradient, despite the absence of the magnetic field. It is thus referred to as the anomalous thermoelectric current. The above result can also be derived using the linear response theory with a precise understanding of the local equilibrium [48].

F. Circular dichroism in ferromagnetic materials

The material response to light is determined by the optical conductivity

$$J = \begin{pmatrix} \sigma_{xx} & \sigma_{xy} \\ -\sigma_{xy} & \sigma_{yy} \end{pmatrix} E . \qquad (91)$$

Here we have assumed that the off-diagonal element only contains the antisymmetric part, which can always be realized by choosing a proper reference frame. At zero frequency, the off-diagonal element reduces to the anomalous Hall conductivity. At finite frequency, we will demonstrate that it can lead to the circular dichroism, referring to the different absorbtion rate of light with opposite helicities.

To derive the circular dichroism, we start with the Maxwell equation for the electric field

$$\nabla \times (\nabla \times E) = -\mu_0 \frac{\partial J}{\partial t} - \frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} , \qquad (92)$$

where μ_0 is the vacuum permeability and c is the speed of light. We can put the following ansatz for the light electric field: $E_0 e^{in\omega/c - i\omega t}$. Then we find that the electric polarization and the refractive index n satisfy the following equation

$$\begin{pmatrix} n^2 - n_0^2 & -ic^2\mu_0\sigma_{xy}/\omega \\ ic^2\mu_0\sigma_{xy}/\omega & n^2 - n_0^2 \end{pmatrix} E = 0 , \qquad (93)$$

where $n_0^2 = 1 + ic^2\mu_0\sigma_{xx}/\omega$. From this, we can solve the eigenvalue of the refractive index

$$n_\pm = \sqrt{n_0^2 \pm c^2\mu_0\sigma_{xy}/\omega} . \qquad (94)$$

For $n_\pm$, the eigenvector reads

$$E_\pm = \begin{pmatrix} 1 \\ \mp i \end{pmatrix} . \qquad (95)$$

As a result, $n_\pm$ corresponds to the refractive index for the right/left circularly polarized light. This indicates that to have different refractive indices for left/right handed light, a nonzero σ_{xy} is required. Using the linear response theory, we can calculate this component of the optical conductivity [67, 68]

$$\sigma_{xy} = -\frac{e^2}{\omega} \sum_{m,n} \int \frac{dk}{(2\pi)^3} \frac{\Delta f_{mn}}{\omega_{mn} + \hbar\omega + i\eta} \text{Im}[(v_x)_{mn}(v_y)_{nm}] , \qquad (96)$$

where $\Delta f_{mn} = f_m - f_n$ and $\omega_{mn} = \varepsilon_m - \varepsilon_n$.

The connection between optical phenomena and the microscopic properties of the ground state is often captured by the sum rule. In the case of the circular dichroism, one can formulate two rules, which is related to the Berry curvature and magnetic moment, respectively. We will assume that in experiments, the refractive index is measurable for left/right handed circularly polarized light through reflection/refraction and absorption. Then according to Eq. (94), the optical conductivity component σ_{xy} can be obtained. The first sum rule directly follows from the Kramers-Kronig relation

$$\begin{aligned} \text{Re}\sigma_{xy}(0) &= \frac{2}{\pi} \int_0^\infty d\omega \frac{\text{Im}\sigma_{xy}}{\omega} \\ &= 2e^2 \int_0^\infty d\omega \sum_{m,n} \frac{\Delta f_{mn}}{\omega^2} \delta(\omega_{mn} + \hbar\omega) \text{Im}[(v_x)_{mn}(v_y)_{nm}] \\ &= \frac{2e^2}{\hbar} \sum_{n>m} \Delta f_{mn} \text{Im}[(\mathcal{A}_x)_{mn}(\mathcal{A}_y)_{nm}] \\ &= -\frac{e^2}{\hbar} \sum_m f_m (\Omega_z)_m . \end{aligned} \qquad (97)$$

Here the subscript $n > m$ means $\varepsilon_n > \varepsilon_m$. One recognizes that the final expression is just the intrinsic anomalous Hall conductivity due to the static electric field.

The second sum rule is the direct integration of $\text{Im}\sigma_{xy}$ [5]

$$\int_0^\infty \text{Im}\sigma_{xy} d\omega$$

$$\begin{aligned} &= 2\pi e^2 \int_0^\infty d\omega \sum_{m,n} \frac{\Delta f_{mn}}{\omega} \delta(\omega_{mn} + \hbar\omega) \text{Im}[(v_x)_{mn}(v_y)_{nm}] \\ &= -2\pi \frac{e^2}{\hbar} \sum_{n>m} \Delta f_{mn} \text{Re}[(\mathcal{A}_x)_{mn}(v_y)_{nm}] \end{aligned}$$

$$= -2\pi \frac{e^2}{\hbar} \sum_{m \in occ}^{n \in unocc} \text{Re}[(\mathcal{A}_x)_{mn}(v_y)_{nm}]$$

$$= -\pi \frac{e^2}{\hbar} \sum_{m \in occ}^{n \in unocc} \text{Re}[(\mathcal{A}_x)_{mn}(v_y)_{nm} - (x \leftrightarrow y)]. \quad (98)$$

Here $m \in occ$ means m runs through the occupied band indices while $n \in unocc$ means that n runs through the unoccupied band indices. By comparing the result to the magnetic moment given in Eq. (41), we immediately find that Eq. (98) is proportional to orbital magnetic moment except that the index n only runs through unoccupied bands.

There is a deeper reason for this difference. We note that although the orbital magnetic moment is invariant under the gauge transformation of the Bloch states, it is not a ground state property. At zero temperature, the ground state density matrix $\rho = \sum_{n \in occ, k} |\psi_{nk}\rangle\langle\psi_{nk}|$ is invariant under any rotation that mixes different occupied bands at the same k-point. Therefore, any ground state property should respect such symmetry. However, the orbital magnetic moment contains the Berry connection $\mathcal{A}_{mn}$ between two occupied bands m and n. As a result, it will change under a rotation of states in the occupied band subspace. On the contrary, the sum rule in Eq. (98) does not change under such a rotation. In this regard, we can claim that the sum of σ_{xy} is proportional to the gauge-invariant part of the orbital magnetic moment.

IV. NONLINEAR TRANSPORT PHENOMENA

In this section we focus on several nonlinear phenomena that are affected by the Berry phase and Berry curvature. In Sect. IV A, we will extend the semiclassical framework up to second order. This lays the groundwork for later discussions. We will discuss some examples in detail to demonstrate the utility of the semiclassical framework and to introduce the new perspective using the geometrical quantities. Specifically, we will discuss the Landau level quantization and the nonlinearity in the Landau level fan diagram measured through the Shubinikov-de Haas oscillations in Sect. IV B. In Sect. IV C, we discuss the field correction to the anomalous Hall current, i.e. the nonlinear anomalous Hall effect. In Sect. IV D and E, we discuss two spatially dispersive phenomena: natural optical activity and nonreciprocal directional dichroism, and demonstrate that they are related to the dipole of magnetic moment and Berry curvature, as well as the quantum metric tensor, respectively.

A. Semiclassical theory up to second order

The semiclassical dynamics in Eq. (57) and (58) cannot fully account for nonlinear phenomena such as the magnetoresistance, magnetoelectric effect, and so on. For this purpose, we need to generalize the semiclassical framework up to second order [69, 70].

Before proceeding to the detail of the theory, we comment that at the second order, there are two distinct perturbations, i.e. the electromagnetic fields to second order and the spatial derivative of the electromagnetic fields. In this section, we focus on the first type and leave the discussion of the second type to the last two parts in this section. From the semiclassical theory point of view, these two types of perturbations can be derived in quite similar manners.

To extend the semiclassical theory up to second order, the construction of the wave packet has to be modified accordingly, to incorporate the modification from the external electromagnetic fields. In fact, the wave packet should be the superposition of the true eigenstates instead of the unperturbed ones. The true eigenstate can be further expanded in the basis of the unperturbed ones, yielding the following wave packet [69]

$$|W\rangle = \int d\boldsymbol{p} e^{i\boldsymbol{p}\cdot\boldsymbol{r}}\left(C_0(\boldsymbol{p})|u_0\rangle + \sum_{n \neq 0} C_n(\boldsymbol{p})|u_n\rangle\right). \quad (99)$$

Here for simplicity we drop the argument $\boldsymbol{p}+(e/\hbar)\boldsymbol{A}(\boldsymbol{r}_c)$ of $|u_0\rangle$ and $|u_n\rangle$. In principle, C_n should be connected to C_0 through perturbation theory. To derive this connection, we still consider the Lagrangian

$$L = \langle W|i\hbar\partial_t - \hat{H}|W\rangle. \quad (100)$$

By taking the variation of L with respect to $C_0^\star$ and $C_n^\star$, we can obtain two sets of equations for the coefficient. This amounts to transforming to the basis spanned by the local unperturbed eigenstates. Specially, the equation through the variation of $C_n^\star$ is equivalent to the following constraint:

$$\langle u_n|e^{-i\boldsymbol{p}\cdot\boldsymbol{r}}(i\hbar\partial_t - \hat{H}_f)|W\rangle = 0, \quad \forall \, n \neq 0. \quad (101)$$

This determines the connection between C_n and C_0.

To calculate it at the first order, we note that

$$\langle u_n|i\hbar\partial_t|W\rangle = \int d\boldsymbol{p}' C_0(\boldsymbol{p}')\langle u_n(\boldsymbol{p})|e^{i(\boldsymbol{p}'-\boldsymbol{p})\cdot\boldsymbol{r}}i\hbar\partial_t|u_0(\boldsymbol{p}')\rangle + i\int d\boldsymbol{p}'\hbar\partial_t C_n(\boldsymbol{p}')\langle u_n(\boldsymbol{p})|e^{i(\boldsymbol{p}'-\boldsymbol{p})\cdot\boldsymbol{r}}|u_n(\boldsymbol{p}')\rangle$$

$$= \frac{e}{2}\boldsymbol{B}\times\dot{\boldsymbol{r}}_c\cdot\mathcal{A}_{n0}C_0(\boldsymbol{p}) + i\hbar\dot{C}_n(\boldsymbol{p})$$

$$= \frac{e}{2}\boldsymbol{B}\times\dot{\boldsymbol{r}}_c\cdot\mathcal{A}_{n0}C_0(\boldsymbol{p}) + \varepsilon_0 C_n(\boldsymbol{p}). \quad (102)$$

In the last equality, we have used that the dynamical phase factor for C_n is $e^{-i\int \varepsilon_0/\hbar dt}$. This is due to two facts: (i) we have

assume that the wave packet is the superposition of the Bloch states from a single band 0 and $|u_n\rangle$ part is just the perturbation;

as a result, it should evolve according to the modified eigenenergy, which is ε_0 at the lowest order; (ii) the time derivative is only on the phase factor of C_n as the derivative on the magnitude of C_n vanishes in the Lagrangian; as a result, we do

not need to concern with the localization of the wave packet around p_c which only affects the magnitude of the coefficients C_0 and C_n.

For the second part, we have

$$
\begin{aligned}
\langle u_n|\hat{H}_0 + \hat{H}_1|W\rangle &= \varepsilon_n C_n + \int d\boldsymbol{p}' C_0(\boldsymbol{p}')\langle u_n(\boldsymbol{p})|e^{i(\boldsymbol{p}'-\boldsymbol{p})\cdot\boldsymbol{r}}\frac{e}{2}\boldsymbol{B}\times(\boldsymbol{r}-\boldsymbol{r}_c)\cdot\hat{\boldsymbol{v}}|u_0(\boldsymbol{p}')\rangle \\
&= \varepsilon_n C_n + \int d\boldsymbol{p}' C_0(\boldsymbol{p}')\langle u_n(\boldsymbol{p})|\frac{e}{2}\boldsymbol{B}\times(i\partial_{\boldsymbol{p}}-\boldsymbol{r}_c)e^{i(\boldsymbol{p}'-\boldsymbol{p})\cdot\boldsymbol{r}}\cdot\hat{\boldsymbol{v}}|u_0(\boldsymbol{p}')\rangle \\
&= \varepsilon_n C_n + \frac{e}{2}\boldsymbol{B}\times(i\partial_{\boldsymbol{p}}-\boldsymbol{r}_c)\cdot[C_0(\boldsymbol{p})\boldsymbol{v}_{n0}] - \frac{e}{2}\boldsymbol{B}\cdot i\langle\partial_{\boldsymbol{p}}u_n|\times\boldsymbol{v}|u_0\rangle C_0(\boldsymbol{p}) \\
&= \varepsilon_n C_n + \frac{e}{2}\boldsymbol{B}\times(i\partial_{\boldsymbol{p}}+\mathcal{A}_0-\boldsymbol{r}_c)C_0(\boldsymbol{p})\cdot\boldsymbol{v}_{n0} - \frac{e}{2}\boldsymbol{B}\cdot\sum_{m\neq 0}\boldsymbol{v}_{nm}\times\mathcal{A}_{m0}\,.
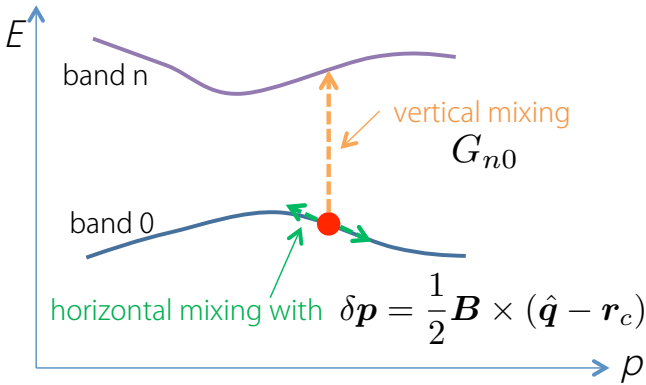\end{aligned} \tag{103}
$$

FIG. 1. The correction to wave packet from magnetic field. $\hat{\boldsymbol{q}}$ has the same meaning of $\boldsymbol{r}$. From Ref. [70].

E — band n — vertical mixing G_{n0} — band 0 — horizontal mixing with $\delta\boldsymbol{p} = \frac{1}{2}\boldsymbol{B}\times(\hat{\boldsymbol{q}}-\boldsymbol{r}_c)$

We can then combine Eq. (102) and (103) and solve the

constraint at the linear order of electromagnetic fields. The result reads

$$
C_n = \frac{G_{n0}}{\varepsilon_0 - \varepsilon_n}C_0 - \frac{i}{2}\frac{e}{\hbar}[\boldsymbol{B}\times(i\partial_{\boldsymbol{p}}+\mathcal{A}_0-\boldsymbol{r}_c)C_0]\cdot\mathcal{A}_{n0}\,, \tag{104}
$$

where $G_{n0} = -\boldsymbol{B}\cdot\boldsymbol{M}_{n0} + e\boldsymbol{E}\cdot\mathcal{A}_{n0}$ with $\boldsymbol{M}_{n0} = \frac{e}{2}\sum_{m\neq 0}(\boldsymbol{v}_{nm} + \boldsymbol{v}_0\delta_{mn})\times\mathcal{A}_{m0}$ being the interband element of the orbital magnetic moment. The contribution from the spin Zeeman energy can be easily added.

Equation (104) has two contributions with different origins. Due to the appearance of the band gap in the denominator, the first term has the conventional form of the perturbation correction. It is hence referred to as the vertical mixing because it mixes Bloch states from different bands but at the same $\boldsymbol{p}$ point in the Brillouin zone. It also contains the nonadiabatic perturbation, due to the fact that the wave packet $|W\rangle$ is time-dependent because of the time-dependence of the argument $\boldsymbol{p} + (e/\hbar)\boldsymbol{A}(\boldsymbol{r}_c)$ of the Bloch state.

In comparison, the second term in C_n modifies the wave packet in the following way:

$$
\begin{aligned}
&\int d\boldsymbol{p}\, e^{i\boldsymbol{p}\cdot\boldsymbol{r}}\frac{-i}{2}\frac{e}{\hbar}[\boldsymbol{B}\times(i\partial_{\boldsymbol{p}}+\mathcal{A}_0-\boldsymbol{r}_c)C_0]\cdot\sum_{n\neq 0}\mathcal{A}_{n0}|u_n\rangle \\
&= \int d\boldsymbol{p}\, e^{i\boldsymbol{p}\cdot\boldsymbol{r}}\frac{e}{2\hbar}[\boldsymbol{B}\times(i\partial_{\boldsymbol{p}}+\mathcal{A}_0-\boldsymbol{r}_c)C_0]\cdot\hat{D}|u_0\rangle \\
&= \int d\boldsymbol{p}\, e^{i\boldsymbol{p}\cdot\boldsymbol{r}}\frac{e}{2\hbar}[\boldsymbol{B}\times(\boldsymbol{r}+\mathcal{A}_0-\boldsymbol{r}_c)C_0]\cdot\hat{D}|u_0\rangle - \int d\boldsymbol{p}\, e^{i\boldsymbol{p}\cdot\boldsymbol{r}}C_0\frac{e}{2\hbar}[\boldsymbol{B}\times i\partial_{\boldsymbol{p}}]\cdot\hat{D}|u_0\rangle \\
&= \int d\boldsymbol{p}\, e^{i\boldsymbol{p}\cdot\boldsymbol{r}}\frac{e}{2\hbar}[\boldsymbol{B}\times(\boldsymbol{r}-\boldsymbol{r}_c)C_0]\cdot\hat{D}|u_0\rangle + \int d\boldsymbol{p}\, e^{i\boldsymbol{p}\cdot\boldsymbol{r}}C_0\frac{e}{2\hbar}\boldsymbol{B}\cdot\boldsymbol{\Omega}|u_0\rangle\,.
\end{aligned} \tag{105}
$$

where $\hat{D} = \partial_{\boldsymbol{p}} + i\mathcal{A}_0$ is the covariant derivative that ensures the gauge invariance. Notice that the momentum argument of $|u_0\rangle$ is $\boldsymbol{p} + (e/\hbar)\boldsymbol{A}(\boldsymbol{r}_c)$. The first term in Eq. (105) has the effect of shifting the momentum to $\boldsymbol{p} + (e/\hbar)\boldsymbol{A}(\boldsymbol{r})$. This is a special property of the perturbation from the magnetic field. It suggests that the correction to the wave function also obeys the Peierls substitution. The same property can also be de-

rived using the Moyal product and phase space formulation of quantum mechanics [71]. The second term in Eq. (105) depends on the Berry curvature and it makes the total correction normal to the original state $e^{i\boldsymbol{p}\cdot\boldsymbol{r}}|u_0\rangle$ to eliminate redundancy in the perturbative correction. The second term in C_n is referred to as the horizontal mixing because it mixes Bloch states in the same band but at neighbouring $\boldsymbol{p}$ points. Both corrections

can be visualized in Fig. 1.

With this new wave packet, the center of mass position in Eq. (37) will change

$$
\begin{aligned}
\boldsymbol{r}_c &= \langle W|\boldsymbol{r}|W\rangle \\
&= \frac{\partial \gamma}{\partial \boldsymbol{p}}\Big|_{\boldsymbol{p}=\boldsymbol{p}_c} + \mathcal{A}_0 \\
&\quad + \int d\boldsymbol{p} C_0^\star(\boldsymbol{p}) C_n(\boldsymbol{p}) \langle u_0|i\partial_{\boldsymbol{p}}|u_n\rangle + c.c..
\end{aligned}
\tag{106}
$$

The last term represents the additional shift to the center of mass position due to the external fields, which we refer to as the positional shift and label it by $\mathcal{A}_0'$. It can be further calculated as follows

$$
\begin{aligned}
\mathcal{A}_0' &= \int d\boldsymbol{p} |C_0|^2 \frac{G_{n0}\mathcal{A}_{0n} + c.c.}{\varepsilon_0 - \varepsilon_n} \\
&\quad - \frac{ie}{2\hbar} \int d\boldsymbol{p} C_0^\star [\boldsymbol{B} \times (\partial_{\boldsymbol{p}}\gamma + \mathcal{A}_0 - \boldsymbol{r}_c) C_0] \cdot \mathcal{A}_{n0}\mathcal{A}_{0n} + c.c. \\
&\quad + \frac{e}{2\hbar} \int d\boldsymbol{p} C_0^\star e^{-i\gamma} \boldsymbol{B} \times \partial_{\boldsymbol{p}}|C_0| \cdot \mathcal{A}_{n0}\mathcal{A}_{0n} + c.c. \\
&= \frac{G_{n0}\mathcal{A}_{0n} + c.c.}{\varepsilon_0 - \varepsilon_n} + \frac{e}{2\hbar}\mathrm{Re} \int d\boldsymbol{p} \boldsymbol{B} \times \partial_{\boldsymbol{p}}|C_0|^2 \cdot \mathcal{A}_{n0}\mathcal{A}_{0n} \\
&= \frac{G_{n0}\mathcal{A}_{0n} + c.c.}{\varepsilon_0 - \varepsilon_n} - \frac{e}{2\hbar}(\boldsymbol{B} \times \partial_{\boldsymbol{p}})_i \cdot \mathrm{Re}[(\mathcal{A}_i)_{n0}\mathcal{A}_{0n}].
\end{aligned}
\tag{107}
$$

It is easy to check that the positional shift is gauge-independent.

The two terms in the positional shift is a direct reflection of the two contributions in the correction to the wave packet state. In fact, the first term in the positional shift is due to the vertical mixing and the second term is due to the horizontal mixing. We find that the horizontal mixing yields a purely geometric correction to the Berry connection. This is not a coincidence, because the horizontal mixing involves neighbouring Bloch states in the momentum space. Their difference is proportional to the Berry connection and their distance is determined by the quantum metric. This positional shift has also been envisioned in Ref. [72], using a similar technique.

Using the modified wave packet, we can derive the Lagrangian. The process is exactly the same with that in the semiclassical theory up to first order. The energetic part will pick up an additional second order correction, and the dynamical part is the same with the expression on the fourth line in Eq. (43). We then use the new expression for the center of mass position and obtain the following Lagrangian

$$
L = -(\boldsymbol{r}_c - \mathcal{A}_0^{\mathrm{t}}) \cdot \hbar\dot{\boldsymbol{k}}_c - \frac{1}{2}e\boldsymbol{B} \times \boldsymbol{r}_c \cdot \dot{\boldsymbol{r}}_c - \tilde{\varepsilon}_0,
\tag{108}
$$

where $\mathcal{A}_0^{\mathrm{t}}$ is the Berry connection evaluated in the true eigen state instead of the unperturbed one. It can be put in the form of the original Berry connection plus the positional shift correction: $\mathcal{A}_0^{\mathrm{t}} = \mathcal{A}_0 + \mathcal{A}_0'$. This Lagrangian has the exact same form with the previous one in Eq. (44). Consequently, the connection between physical and canonical variables also has the same form except that the Berry connection $\mathcal{A}_0^{\mathrm{t}}$ in the exact eigenstate should be used

$$
\boldsymbol{r}_c = \boldsymbol{q} + \mathcal{A}_0^{\mathrm{t}} + \frac{e}{2\hbar}(\boldsymbol{B} \times \mathcal{A}_0^{\mathrm{t}} \cdot \partial_{\boldsymbol{p}})\mathcal{A}_0^{\mathrm{t}} + \frac{e}{2\hbar}\Omega_0^{\mathrm{t}} \times (\boldsymbol{B} \times \mathcal{A}_0^{\mathrm{t}}),
\tag{109}
$$

$$
\boldsymbol{k}_c = \boldsymbol{p} + \frac{e}{2\hbar}\boldsymbol{B} \times \boldsymbol{q} + \frac{e}{\hbar}\boldsymbol{B} \times (\boldsymbol{r}_c - \boldsymbol{q}),
\tag{110}
$$

where $\Omega_0^{\mathrm{t}} = \nabla_{\boldsymbol{p}} \times \mathcal{A}_0^{\mathrm{t}}$ is the Berry curvature evaluated using the exact eigenstate. This suggests the following phase space density of states

$$
\mathcal{D} = 1 + \frac{e}{\hbar}\boldsymbol{B} \cdot \Omega_0^{\mathrm{t}}.
\tag{111}
$$

One can also derive the equations of motion up to second order using the variational principle. Interestingly, they keep the same form [69]

$$
\dot{\boldsymbol{r}}_c = \frac{\partial \tilde{\varepsilon}_0}{\hbar\partial \boldsymbol{k}_c} - \dot{\boldsymbol{k}}_c \times \Omega_0^{\mathrm{t}}(\boldsymbol{k}_c),
\tag{112}
$$

$$
\hbar\dot{\boldsymbol{k}}_c = -e\boldsymbol{E} - e\dot{\boldsymbol{r}}_c \times \boldsymbol{B}.
\tag{113}
$$

Finally, we comment that as the essential ingredient in the semiclassical dynamics up to the second order, the positional shift has the meaning of the change in the electric dipole moment of the wave packet. It can be used to derive the polarizability. For this purpose, we apply external electromagnetic fields, and there will be two changes in the electric polarization accordingly. First, the density of states has to be changed to $\mathcal{D}$. Second, the Berry connection $\mathcal{A}_0 \rightarrow \mathcal{A}_0 + \frac{e}{2\hbar}(\boldsymbol{B} \times \mathcal{A}_0 \cdot \partial_{\boldsymbol{p}})\mathcal{A}_0 + \frac{e}{2\hbar}\Omega_0^{\mathrm{t}} \times (\boldsymbol{B} \times \mathcal{A}_0)$ according to Eq. (109). Adding up these two modifications, we find that the first order change in electric polarization is

$$
\delta P = -e \int \frac{d\boldsymbol{k}}{(2\pi)^3} \left[\frac{e}{2\hbar}(\Omega_0 \cdot \mathcal{A}_0)\boldsymbol{B} + \mathcal{A}_0' \right].
\tag{114}
$$

The first term is the Abelian Chern-Simons 3-form, which corresponds to the topological part of the orbital magnetoelectric coefficient. The second term can give rise to both the electric polarizability and the cross-gap part of the orbital magneto-electric polarizability, consistent with the calculation using the linear response theory [73]. This confirms the validity of the first order correction to the Berry connection.

B. Landau level quantization and nonlinear Landau level fan diagram

The first nonlinear phenomena that we explore is the non-linearity in the Landau level fan diagram measured through the quantum oscillation experiment. Landau levels in solids are described by the Onsager's rule [74]

$$
S(\varepsilon_n) = 2\pi\left(n + \frac{1}{2}\right)\frac{eB}{\hbar},
\tag{115}
$$

where $S(\varepsilon_n)$ is the area in the momentum space enclosed by an equal-energy contour. If we plot the level index as a function of $1/B$, all data points should fall on the straight lines

$$
n = \frac{\hbar S}{2\pi eB} - \frac{1}{2}.
\tag{116}
$$

The slope of the line is governed by the area S, which can be tuned by changing the quantization energy through, for example, gating. This type of pattern forms the Landau level fan diagram.
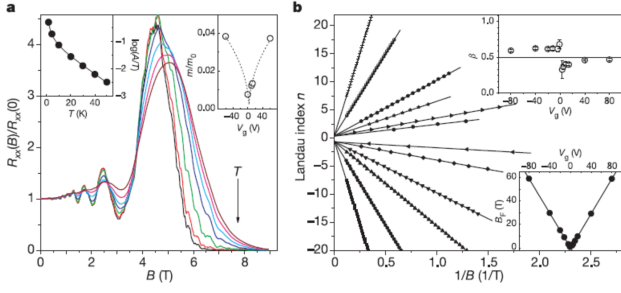
FIG. 2. Temperature-dependence of the Shubnikov-de Haas oscil-
lations (a) and the gate-voltage-dependence of the Landau level fan
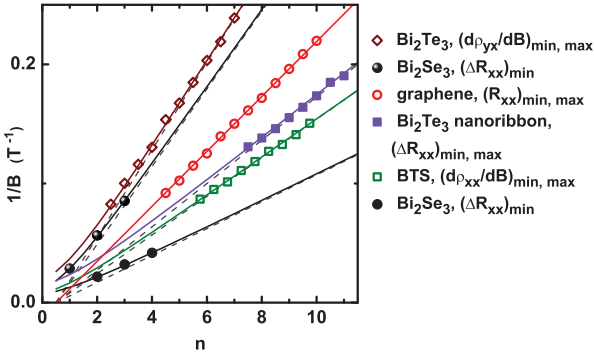diagram (b) in graphene. From Ref. [81].

FIG. 3. Landau level fan diagrams observed in various topological
insulators and graphene. The nonlinearity is clear at large magnetic
field. From Ref. [82].

As the Berry phase can modify the Lagrangian, it can also
change the Landau level position. In the presence of the Berry
phase, it has been shown that the quantization rule changes
to [75–77]

$$S(\varepsilon_n) = 2\pi \left(n + \frac{1}{2} - \frac{\Gamma}{2\pi} \right) \frac{eB}{\hbar}, \qquad (117)$$

where Γ is the Berry phase associated with the equal-energy
contour in the momentum space. Later this quantization rule
has also been generalized to include the contribution from the
magnetic moment [3, 78, 79] and to include the non-Abelian
case [80].

The role of the Berry phase is most clearly seen in the two-
dimensional gapless Dirac model, as it has a constant Berry
phase π and $-\pi$ for the conduction and valence band. This
will shift the Landau level index by ± 0.5 and hence change
the position of the Landau level. The linear relation between
n and $1/B$ still holds and one can obtain the Berry phase by
fitting the Landau level fan diagram according to Eq. (116) and
calculate the offset in the Landau level index. This has
already been done in the quantum oscillation experiments in
graphene [81, 83], as shown in Fig. 2. However, later quan-
tum oscillation experiments in the surface states of three-
dimensional topological insulators demonstrate violations of
the linearity between n and $1/B$ [82, 84–91], as shown in
Fig. 3. To explain this nonlinearity, one needs a more accu-
rate Landau level quantization rule.

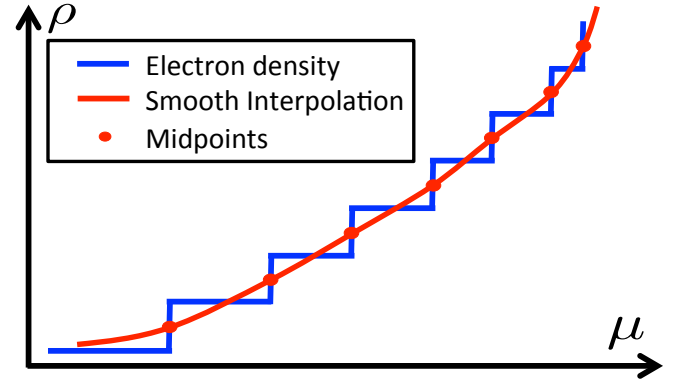To further generalize the Onsager's rule, we assume that

FIG. 4. Density quantization rule for Landau levels. The true elec-
tron density and the semiclassical one are in the blue and red lines,
respectively. From Ref. [92].

instead of the Berry phase, we should have a function of en-
ergy which admits an asymptotic expansion with respect to the
magnetic field, i.e.

$$\left(n + \frac{1}{2} \right) \frac{eB}{h} = \sum_{m=0}^{N} R_m(\varepsilon_n) \frac{B^m}{m!} + O(B^{N+1}). \qquad (118)$$

This meaning of the assumption will be made clear later. By
comparing it with Eq. (117), we should have $R_0 = S/4\pi$ and
$R_1 = e\Gamma/2\pi h$. In general, there is a connection between the co-
efficient R_m with the magnetic response function at zero field
and zero temperature, i.e.

$$R_m = \lim_{T \to 0} \lim_{B \to 0} \frac{\partial^m \rho(B, T, \mu)}{\partial B^m}, \qquad (119)$$

where $\rho(B, T, \mu)$ is the electron density for the Landau level
spectrum.

This relation can be justified as follows. Without loss of
generality, we consider the electron density for Landau level
spectrum $\varepsilon_n(B)$ from a band minimum in two dimensions:

$$\rho(B, T, \mu) = (B/\phi_0) \sum_n f[\varepsilon_n(B) - \mu], \qquad (120)$$

where $\phi_0 = h/e$ is the flux quantum, f is the Fermi function,
and μ is the chemical potential. At $T = 0$, the electron density
is a staircase function with constant risers B/ϕ_0 located at $\mu =
\varepsilon_n(B)$, as shown by the blue curve in Fig.(4).

We then perform a smooth interpolation of the spectrum
$\varepsilon_n(B)$ as follows. We can re-write the spectrum as $\varepsilon_n(B) =
g(x_n, B)$, where $x_n = (n + 1/2)B/\phi_0$ is the zero-temperature
electron density when the n-th Landau level is half-filled. We
then allow x to change continuously, the function $\varepsilon = g(x, B)$
smoothly interpolates the spectrum $\varepsilon_n(B)$ at $x = x_n$ for each
value of B. As $\varepsilon_n(B)$ increases with n, $g(x, B)$ is a mono-
tonic function of x for each B. $g(x, B)$ can be inverted to yield
$x = x(\mu, B)$, represented as the smooth curve in Fig.(4). We
assume that this function $x(\mu, B)$ admits an asymptotic expan-
sion with respect to B, which is actually Eq. (118). By con-
struction, the interpolation condition dictates that this smooth
curve $x(\mu, B)$ crosses midpoints of the staircase risers, i.e.
$x_n = x(\varepsilon_n, B)$. This completes half of the quantization rule,

i.e. the quantized part. We still need to connect the right hand side $x(\mu, B)$ with the property of the system without magnetic field.

We now analyze the right hand side of Eq.(119). The order of the limit $T \to 0$ and $B \to 0$ can be realized in the following scenario: Landau level spacing is much smaller than $k_B T$, and $k_B T$ is much smaller than the inherent energy scale of the band structure. Examples of the latter include the distance between the chemical potential and band singularity energy such as the band bottom and band top). The first condition validates the use of the Euler-Maclaurin formula to express the summation over n in Eq. (120) as an integration over a continuous variable x:

$$\rho(B, T, \mu) = \int_{B/2\phi_0}^{\infty} f \, dx + \frac{Bf|_{x=B/2\phi_0}}{2\phi_0} + \mathcal{R}, \quad (121)$$

where $f = f[g(x, B) - \mu]$. $\mathcal{R}$ contains all the remainder terms.

$\mathcal{R}$ only involves exponentially small terms at low temperature. Terms in $\mathcal{R}$ should be calculated at $x = B/2\phi_0$, i.e. at the 0-th Landau level near the band minimum. We have ignored the contribution at $x = \infty$ since they correspond to very high energy and hence are exponentially small as $T \to 0$. The arguments can be easily generalized for realistic models in solid state physics which contain singular points such as the van Hove singularities. Moreover, terms in $\mathcal{R}$ are proportional to successively higher power of B and contain $\partial f / \partial \mu$ or its higher order derivatives with respect to μ. Therefore, each term is exponentially small as $T \to 0$.

We now calculate the right hand side of Eq.(119). Due to the above property of $\mathcal{R}$, only finite number of terms in $\mathcal{R}$ contribute to $\lim_{B \to 0}(\partial^m \rho / \partial B^m)$. We then take the limit $T \to 0$. The first two terms in Eq.(121) will yield x, and finite number of terms from $\mathcal{R}$ vanishes. As a result, $\lim_{T \to 0} \lim_{B \to 0}(\partial^m \rho / \partial B^m) = (\partial^m x / \partial B^m)|_{B=0}$. Under the limit that $k_B T$ is much larger than the Landau level spacing, the true electron density can be approximated by the semiclassical one ρ_{semi}, and the latter is exactly the meaning of x.

In the following, we prove the identity $R_m = (\partial^m x / \partial B^m)|_{B=0}$. This will be able to establish the equality in Eq.(119). Note that the left hand side of Eq.(118) is $x_n = (n + 1/2)B/\phi_0$. Therefore, if ε_n in the function $R_m(\varepsilon_n)$ on the right hand side of Eq.(118) is replaced by a continuous variable μ, we get a continuous function $x = x'(\mu, B)$. If this quantization rule in Eq. (118) works, i.e. $x_n = x'(\varepsilon_n, B)$, this function x' has to coincide with another function directly derived from the Landau level spectrum, i.e. $x = x(\mu, B)$. Therefore, $R_m = (\partial^m x' / \partial B^m)|_{B=0} = (\partial^m x / \partial B^m)|_{B=0}$. This completes the proof of Eq.(119).

Based on the above discussions, the quantization rule in Eq. (118) can also be concisely represented by the following density quantization rule [92]

$$\rho_{\text{semi}}(\varepsilon_n) = \left(n + \frac{1}{2}\right) \frac{eB}{h}. \quad (122)$$

This suggests that the smooth semiclassical electron density always intersects with the true stepwise electron density at the half-filling points, as shown in Fig. 4.

One can connect R_m in Eq.(118) with the familiar magnetic response functions derived from the free energy, such as the magnetization M and the magnetic susceptibility χ at zero temperature. The semiclassical electron density is related to the semiclassical free energy: $\rho_{\text{semi}}(B, T, \mu) = -(\partial/\partial\mu)G_{\text{semi}}(B, T, \mu)$. The semiclassical free energy can be expanded with respect to B: $G_{\text{semi}} = G_0 - BM - \chi B^2/2 + \cdots$. Therefore, we can identify the following relation: $R_0 = S/(4\pi^2)$, $R_1 = \partial M/\partial\mu$, $R_2 = \partial\chi/\partial\mu$, and so on. Here S is the k-space area enclosed by the equal energy contour, and proportional to the semiclassical electron density R_0 at $B = 0$. The zero-field magnetic response functions up to second order, i.e. M and χ, have been studied using both the linear response theory and the semiclassical theory [38, 47, 93–95].

With the expression of the coefficient we can discuss the connection between the new quantization rule in Eq. (118) and the previous Onsager's rule. Truncating the quantization rule in Eq. (118) at zeroth order recovers the Onsager's rule: from the expression of R_0, we can obtain $S_0 = 2\pi\left(n + \frac{1}{2}\right)\frac{eB}{\hbar}$. Truncating Eq.(118) at first order can recover the well-known Berry phase and magnetic moment correction to the Onsager's rule. The magnetization is given by $M = \int(mf - \Omega g)d^2k/(4\pi^2)$. Therefore, $\frac{\partial M}{\partial\mu} = -\int(mf' - \Omega f)\frac{d^2k}{4\pi^2}$. If we combine the first term with R_0 and move the second term to the left hand side of Eq.(118), we obtain the following quantization condition: $S' = 2\pi\left(n + \frac{1}{2} - \frac{\Gamma(\mu)}{2\pi}\right)\frac{eB}{\hbar}$, where $S' = \int f(\varepsilon_0 - Bm - \mu)d^2k$ is the area enclosed by the equal-energy contour in the modified band structure $\varepsilon_0 - Bm$ with the energy μ, and $\Gamma(\mu)$ is the Berry phase associated with the semiclassical orbit. This is exactly the Onsager's rule with the Berry phase and magnetic moment modification [3, 75, 77, 78].

Truncating the quantization rule at second order, we have

$$\left(n + \frac{1}{2}\right)\frac{eB}{h} = \frac{S(\varepsilon_n)}{4\pi^2} + B \left.\frac{\partial M}{\partial\mu}\right|_{\mu=\varepsilon_n} + \frac{1}{2}B^2 \left.\frac{\partial\chi}{\partial\mu}\right|_{\mu=\varepsilon_n}. \quad (123)$$

It is interesting to note that Eq. (123) indicates the nonlinearity in the Landau level fan diagram when plotted as n against $1/B$ due to the appearance of the susceptibility. Such nonlinearity can account for that observed in experiments [85–91], as shown later.

As a concrete example, we consider the following model that describes surface states of three dimensional topological insulators [82] (for simplicity, we choose e, $\hbar$ to be unity):

$$\hat{H} = v_f(k_x\sigma_y - k_y\sigma_x) + \frac{k^2}{2m_{\text{eff}}}, \quad (124)$$

where v_f is the Fermi velocity, m_{eff} is the effective mass, and σ are Pauli matrices in the spin space. The three terms in Eq.(124) represents spin-orbit coupling, kinetic energy, and spin Zeeman energy, respectively. Under a B field along z direction, Landau levels in the conduction band reads: $\varepsilon_{\text{quan}} = nB/m_{\text{eff}} + \sqrt{2v_f^2 nB + (m_0/m_{\text{eff}} - g_s/2)^2\mu_B^2 B^2}$, where m_0 is the free electron mass, g_s is the surface g-factor, and μ_B is the Bohr magneton.

We first discuss the simple case when $1/m_{\text{eff}} \to 0$ and $g_s = 0$. Equation (124) then reduces to a perfect two-dimensional Dirac model, with π Berry phase for the conduction band and vanishing magnetic moment. In this case, the quantization rule in Eq.(123) up to linear order on the right hand side yields the exact Landau levels $\varepsilon_{\text{quan}} = v_f\sqrt{2nB}$. This coincidence suggests that $R_\ell = 0$ with $\ell \geq 2$ for this model, when μ falls inside the band. Specifically, at $\ell = 2$, the vanishing $\partial\chi/\partial\mu$ when μ falls inside the band has already been confirmed in previous literature [93–95].
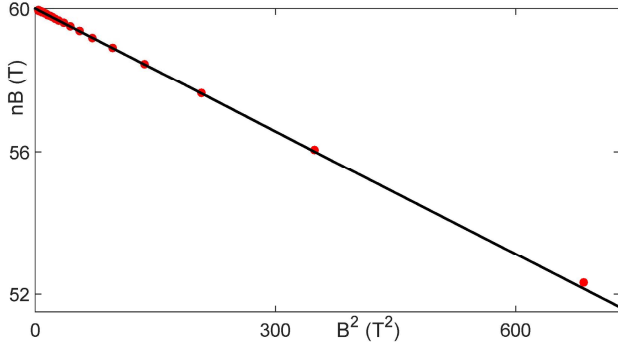
FIG. 5. Comparing exact spectrum with Eq.(126). The black line stands for the exact spectrum and the red dots stand for the quantization rule. From Ref. [92].

Now we discuss the general case with a finite m_{eff} and g_s. In this case, the susceptibility does not necessarily vanish. In fact, when μ falls inside the conduction band, the susceptibility reads

$$\frac{\partial \chi}{\partial \mu} = -\frac{m_{\text{eff}} \mu_B^2}{2\pi v_f} \frac{\left(\frac{1}{m_{\text{eff}}/m_0} - \frac{1}{2}g_s\right)^2}{k_f + m_{\text{eff}} v_f}, \qquad (125)$$

where $k_f = -m_{\text{eff}} v_f + \sqrt{m_{\text{eff}}^2 v_f^2 + 2m_{\text{eff}}\mu}$ is the Fermi wave vector. The Berry phase is still π and the magnetic moment still vanishes. Therefore, Eq.(123) yields

$$n = k_f^2/2B - \frac{m_{\text{eff}} B \mu_B^2}{2v_f} \frac{\left(\frac{1}{m_{\text{eff}}/m_0} - \frac{1}{2}g_s\right)^2}{k_f + m_{\text{eff}} v_f}. \qquad (126)$$

Due to the appearance of the second term in Eq.(126), n does not depend on $1/B$ linearly when $m_0/m_{\text{eff}} - g_s/2$ does not vanish.

We can compare the exact spectrum with the quantization rule in Eq.(126) in a nB-versus-B^2 plot which may be more helpful to experiments, as shown in Fig.(5). We first note that if n linearly depends on $1/B$, nB would be a constant. Therefore, the inclination of the exact spectrum data clearly suggests a nonlinear relation. According to Eq.(126), the π Berry phase is implied by the straightness of the solid line. The slope of the line yields $\partial \chi / \partial \mu$, and its interception to the y axis gives the Fermi surface area. As shown in Fig.(5), the exact spectrum fits Eq.(126) quite well. As experimental data fits the exact Landau level spectrum from Eq.(124) quite well, from Fig. (5), it will also fit the quantization rule in Eq.(126), demonstrating its utility.

C. Nonlinear anomalous Hall current

As discussed in Sect. IV D, the anomalous Hall effect requires broken time-reversal symmetry as well as certain mirror symmetries. This can be realized by either a net spin magnetization as in ferromagnets or an orbital magnetization as in

noncolinear antiferromagnets. It is forbidden if time reversal symmetry is present. However, the next order contribution to the Hall effect, i.e. the nonlinear Hall effect, may still be present, because a net magnetization can be induced by electromagnetic fields.

We first briefly introduce the general framework of deriving a current in the semiclassical theory. The current in the semiclassical theory reads

$$\boldsymbol{J} = -e \int \frac{d\boldsymbol{k}}{8\pi^3} \mathcal{D}\dot{\boldsymbol{r}} f, \qquad (127)$$

where f is the electron distribution function, which is the Fermi function in equilibrium. Here and hereafter, we drop the subscript c in the center of mass position $\boldsymbol{r}_c$ and momentum $\boldsymbol{k}_c$ for simplicity. By plugging the force in Eq. (113) into the velocity in Eq. (112), we obtain

$$\mathcal{D}\dot{\boldsymbol{r}} = \tilde{\boldsymbol{v}} + e\boldsymbol{E} \times \boldsymbol{\Omega}^{\text{t}} + \frac{e}{\hbar}\left(\tilde{\boldsymbol{v}} \cdot \boldsymbol{\Omega}^{\text{t}}\right)\boldsymbol{B}, \qquad (128)$$

where $\tilde{\boldsymbol{v}} = \partial \tilde{\varepsilon}/\hbar \partial \boldsymbol{k}$ is the modified band velocity. Here and hereafter, we will ignore the band index 0 for simplicity and the subscript 0 only has the meaning of zeroth order.

The distribution function f in Eq. (127) can be solved from the Boltzmann equation. Under the relaxation time approximation, the Boltzmann equation reads

$$\dot{\boldsymbol{k}} \cdot \frac{\partial f}{\partial \boldsymbol{k}} = \left.\frac{df}{dt}\right|_{\text{collision}} = -\frac{f - f_0}{\tau}, \qquad (129)$$

where τ is the relaxation time. We emphasize that the argument of equilibrium distribution f_0 is the modified band energy $\tilde{\varepsilon}$. Solving the Boltzmann equation perturbatively, one has [96]

$$f = \sum_{m=0}^{\infty} (-\tau \dot{\boldsymbol{k}} \cdot \boldsymbol{\partial}_k)^m f_0(\tilde{\varepsilon}). \qquad (130)$$

This completes the theory for current in the semiclassical framework.

Here for the purpose of deriving the correction to the anomalous Hall effect, we will only keep the solution in Eq. (130) up to first order. The result reads

$$f_1 = \frac{e\tau}{\hbar} \boldsymbol{E} \cdot \boldsymbol{v}_0 f_0', \qquad (131)$$

where $f_0' = \frac{\partial f_0}{\partial \varepsilon}$.

We can then discuss the anomalous Hall current derived in the semiclassical theory. We first consider the contribution equilibrium distribution f_0. According to Eq. (128), we have the following contribution

$$\boldsymbol{J}_1 = -e \int \frac{d\boldsymbol{k}}{8\pi^3} [\tilde{\boldsymbol{v}} + e\boldsymbol{E} \times \boldsymbol{\Omega}^{\text{t}}] f_0(\tilde{\varepsilon}). \qquad (132)$$

It is easy to show that the first term vanishes in general and we have

$$\boldsymbol{J}_1 = -e^2 \boldsymbol{E} \times \int \frac{d\boldsymbol{k}}{8\pi^3} \boldsymbol{\Omega}^{\text{t}} f_0(\tilde{\varepsilon}) \qquad (133)$$

At lowest order, this will give rise to the anomalous Hall current discussed previously. At second order, due to the field
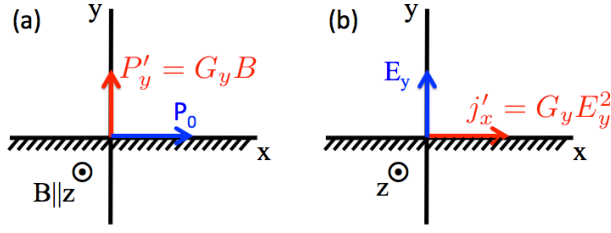
FIG. 6. Magnetoelectric effect (Panel a) and the related nonlinear anomalous Hall induced by the electric field (Panel b). From Ref. [69].

correction to both the Berry curvature and band energy, we have nonlinear anomalous Hall current.

We first consider systems with both time reversal and inversion symmetry. In this case, a net magnetization can be induced through the magnetic susceptibility by a magnetic field, hence leading to a Hall-type current. The analytical expression for this correction can be obtained by keeping the magnetic-field-dependent part in $\mathbf{\Omega}^{t}$ and the mangetic-field-correction to the band energy in Eq. (133). The result reads [69]

$$ \boldsymbol{J} = \frac{e^2}{\hbar}\boldsymbol{E} \times \int [\hbar\boldsymbol{v} \times \mathcal{A}'(\boldsymbol{B}) + \boldsymbol{\Omega}(\boldsymbol{B} \cdot \boldsymbol{m})]f_0' \frac{d\boldsymbol{k}}{8\pi^3} , \quad (134) $$

where $\mathcal{A}'(\boldsymbol{B})$ stands for the part of $\mathcal{A}'$ solely dependent on $\boldsymbol{B}$.

We note that the ordinary Hall current is also linear in electric field and magnetic field. However, it has different origins from the one in Eq. (134). The ordinary Hall current is due to the Lorentz force and the second order solution to the distribution function in the Boltzmann equation. In comparison, the current in Eq. (134) is due to a nontrivial geometric structure (Berry curvature, magnetic moment and the quantum metric tensor) in the momentum space and is independent of the relaxation time. This difference can be most easily seen through the ratio of the resistivity ρ_{xy}' from Eq. (134) and the ordinary Hall resistivity ρ_{xy}^{ord}

$$ \frac{\rho_{xy}'}{\rho_{xy}^{\mathrm{ord}}} = \left(\rho_{xx} \frac{e^2}{4h} \right)^2 S(\mu) , \quad (135) $$

where the first factor reflects the different dependence on the relaxation time and is universal, and $S(\mu)$ is a model-

dependent factor but independent of the relaxation time. when, In dirty metals/semiconductors with relatively small relaxation time, ρ_{xx} is large, and ρ_{xy}' will dominate the ordinary Hall effect. In a typical Hall-bar measurement set-up, both ρ_{xy}' and ρ_{xy}^{ord} should contribute to the Hall current. They should be differentiated by changing the universal scaling factor through temperature, film thickness, or doping, and measure the scaling behaviour based on Eq. (135).

We now consider systems that simultaneously break the time reversal and inversion symmetry but preserve the combined symmetry. This symmetry allows the magnetoelectric effect, such that a magnetization can be induced by an electric field through the magnetoelectric coefficient [97]. This induced magnetization can further lead to a Hall-type current. The analytical expression can be obtained by keeping the electric-field-dependent part in the Berry curvature in Eq. (133). The result reads [69]

$$ \boldsymbol{J} = e^2 \boldsymbol{E} \times \int [\boldsymbol{v} \times \mathcal{A}'(\boldsymbol{E})]f_0' \frac{d\boldsymbol{k}}{8\pi^3} , \quad (136) $$

where $\mathcal{A}'(\boldsymbol{E})$ stands for the part of $\mathcal{A}'$ solely dependent on $\boldsymbol{E}$. This current is connected to the magnetoelectric effect, as shown in Fig. 6. The mirror-y symmetry can forbid the anomalous Hall current but not this nonlinear anomalous Hall current, i.e. by going from the linear to nonlinear anomalous Hall current, although the time-reversal symmetry still needs to be broken, we can relax the constraint on the mirror symmetry.

As a concrete example, we consider the following two-band toy model

$$ \hat{H} = v'k_x + vk_x\sigma_x\tau_y + vk_y\sigma_y + \Delta\sigma_z , \quad (137) $$

where $\boldsymbol{\sigma}$ stands for the pseudospin in the orbital space, τ_y is the spin Pauli matrix, v' and v are two velocity parameters, and Δ gives a finite band gap. The energy spectrum of this model is $\varepsilon_{\pm} = v'k_x \pm \varepsilon$ with $\varepsilon = \sqrt{v^2k_x^2 + v^2k_y^2 + \Delta^2}$. When $v' = 0$, this model describes a two-dimensional Dirac model. The v' adds a tilting to the Dirac cone. The spin component τ_y breaks mirror-x and mirror-z symmetry but respects mirror-y symmetry. This will forbid the anomalous Hall current in the xy plane.

For the nonlinear anomalous Hall current, according to Fig. 6, we apply the electric field along y direction. We also assume that the Fermi energy falls in the conduction band. Then the current in Eq. (136) becomes

$$ J_x = 2e^3 E_y^2 \mathrm{Re} \int \frac{v_x(\mathcal{A}_y)_{0n}(\mathcal{A}_y)_{n0} - v_y(\mathcal{A}_x)_{0n}(\mathcal{A}_y)_{n0}}{\varepsilon_0 - \varepsilon_n} f_0' \frac{d\boldsymbol{k}}{4\pi^2} . \quad (138) $$

We can use the toy model in Eq. (137) to calculate the above current. The result reads

$$ J_x = v^2 e^3 E_y^2 \int \frac{d\boldsymbol{k}}{4\pi^2} \frac{1}{4\varepsilon^3} \left[v' \left(1 - \frac{v^2k_y^2}{\varepsilon^2} \right) + \frac{v^2k_x}{\varepsilon} \right] f_0' . \quad (139) $$

In the limit that $\Delta = 0$, the above integration can be easily

calculated:

$$ \begin{aligned} J_x &= \frac{v^3 e^3 E_y^2}{16\pi^2\mu^3} \int d\theta \left(1 + \frac{v'}{v}\cos\theta \right)^2 \left(\cos\theta + \frac{v'}{v}\cos^3\theta \right) \\ &= \frac{v'v^2 e^3 E_y^2}{8\pi^2\mu^3} \int d\theta \left(\cos^2\theta + \frac{v'}{v}\cos^4\theta \right) \end{aligned} $$

$$= \frac{v'v^2 e^3 E_y^2}{4\pi^2 \mu^3}\left(1 + \frac{3v'}{8v}\right). \tag{140}$$

This clearly demonstrate the existence of the nonlinear anomalous Hall current. We can also find that the tilting plays an important role. In fact, if we set $v' = 0$, the nonlinear anomalous Hall current vanishes. This current also decays quickly to zero when the chemical potential moves away from the Dirac point.

We now consider the nonlinear anomalous Hall current due to the contribution from f_1. The current reads

$$\boldsymbol{J}_2 = -\frac{e^2\tau}{\hbar}\int\frac{d\boldsymbol{k}}{8\pi^3}(\boldsymbol{v}_0 + e\boldsymbol{E}\times\boldsymbol{\Omega}^{\mathrm{t}})\boldsymbol{E}\cdot\boldsymbol{v}_0 f_0'. \tag{141}$$

It is easy the check that the first term in the bracket does not contribute to an antisymmetric component of the conductivity. Therefore, we are left with

$$\boldsymbol{J}_2 = -\frac{e^3\tau}{\hbar}\boldsymbol{E}\times\int\frac{d\boldsymbol{k}}{8\pi^3}\boldsymbol{\Omega}^{\mathrm{t}}\boldsymbol{E}\cdot\boldsymbol{v}_0 f_0'. \tag{142}$$

Due to the transformation property of $\boldsymbol{\Omega}$ and $\boldsymbol{v}_0$, this current can appear in noncentrosymmetric but time-reversal-invariant materials. It is due to the net magnetization induced in the nonequilibrium steady state through the Edelstein effect. Interestingly, the requirement of the mirror symmetry in Fig. 6(b) also works for this current. But one should keep in mind that for the current in Eq. (136), as the time reversal symmetry is broken, the inherent spin texture in the sample is also subject to the mirror operation. The observation of this nonlinear Hall current has been reported in several recent experiments [10–12]. We comment that in Eq. (142), $\boldsymbol{v}\boldsymbol{\Omega}$ together constitutes a pseudotensor which represents the first order moment of the Berry curvature in the momentum space. Hence it is referred to as the Berry curvature dipole.

A concrete example of this nonlinear anomalous Hall current is provided in Ref. [9]. At low temperature, the surface state of the three-dimensional topological insulator can enter into a ferroelectric state which meets the symmetry requirement. The model Hamiltonian is given by

$$\hat{H} = v_x k_x \sigma_y - s v_y k_y \sigma_x + s\alpha k_y + \beta\sigma_z, \tag{143}$$

where $s = \pm 1$, β opens a band gap, and α tilts the Dirac cone. This model breaks the mirror-x symmetry but keeps the mirror-y symmetry. As a result, the nonlinear anomalous Hall current along x direction should exist, when the electric field is applied along y direction, i.e. we should have

$$J_x = -\frac{e^3\tau}{\hbar}E_y^2\int\frac{d\boldsymbol{k}}{4\pi^2}\Omega_z v_y f_0'. \tag{144}$$

For the conduction band, the Berry curvature reads

$$\Omega_z = \frac{1}{2}\frac{s v_x v_y \beta}{(\beta^2 + v_x^2 k_x^2 + v_y^2 k_y^2)^{3/2}}. \tag{145}$$

For $s = \pm 1$, the Berry curvature has the same magnitude but opposite signs. However, the tilting is also a odd function of s. This indicates that $v_y\Omega_z$ is an even function of s and hence can be nonzero. The final result is

$$J_x = \frac{e^3\tau}{\hbar}E_y^2\frac{3v_x v_y n\beta\alpha\mu(1 + u^2)}{[\mu^2(1 + u^2)(1 + 2u^2) - u^2\beta^2]^{5/2}}, \tag{146}$$

where $u = \alpha'/\sqrt{v^2 - \alpha'^2}$, $\alpha' = \alpha\sqrt{v_x/v_y}$, $n = \gamma^2/(4\pi v\sqrt{v^2 - \alpha'^2})$, and $\gamma = \sqrt{\mu^2 + \mu^2\alpha'^2/(v^2 - \alpha'^2) - \beta^2}$. From this expression, we can find that both the band gap and the tilting are important for the nonlinear anomalous Hall current. The former will make the Berry curvature nonzero, while the latter makes the Berry curvature has a dipolar structure in the momentum space. Moreover, right at the conduction band bottom, $\gamma = 0$ and so is the current J_x. When the chemical potential is high in the conduction band, J_x goes to zero as $1/\mu^2$. As a result, the current J_x as a function of the chemical potential should start at zero, reach a peak, and then fall to zero as $1/\mu^2$.

Finally, we comment that if the time reversal and inversion symmetry, as well as their combined symmetry are all broken, the anomalous Hall and its two nonlinear versions in Eq. (136) and (142) should all be present. Since they depend on different band properties, they may have maximum at different chemical potentials, indicating potential tuning possibility for linear/nonlinear behaviour in the anomalous Hall current. Moreover, there are methods to distinguish them. One can first perform a scaling of the relaxation time to distinguish the intrinsic contribution (i.e. independent of τ) from the extrinsic contribution (i.e. linear in τ) in Eq. (142). This is similar to extracting intrinsic, skew-scattering and side-jump contribution from the total anomalous Hall signal [98–103]. The nonlinear part can be further extracted by reversing the direction of the electric field and finding the difference in the Hall signal.

D. Spatially dispersive phenomenon: circular dichroism in noncentrosymmetric materials

As discussed in Sect. III F, in ferromagnetic materials, the left/right handed light is responded differently. In reality, there is a large family of materials that shares the same optical properties, but that breaks the inversion symmetry instead of the time reversal symmetry, such as quartz, RNA and DNA molecules, etc. In such materials, σ_{xy} vanishes identically as dictated by the time reversal symmetry. Their unique optical response to the light chirality is ascribed to the first order correction to σ_{xy} due to the coupling between the light wave vector to the anisotropy of the material in space [97].

In general, the light wave length is much larger than the unit cell length scale. As a result, the light wave vector is much smaller than the crystal momentum. In this regard, we can expand the optical conductivity with respect to the light wave vector $\boldsymbol{q}$:

$$\sigma_{ij}(\omega, \boldsymbol{q}) = \sigma_{ij}(\omega, 0) + \sigma_{ijk}(\omega, 0)q_k + \cdots. \tag{147}$$

Without loss of generality, we assume that the light propagates along z direction. In noncentrosymmetric materials, the optical current is given by

$$\boldsymbol{J} = \begin{pmatrix} \sigma_{xx} & \sigma_{xyz}q_z \\ -\sigma_{xyz}q_z & \sigma_{yy} \end{pmatrix}\boldsymbol{E}. \tag{148}$$

The equation for the refractive index and the electric field component will satisfy

$$\begin{pmatrix} n^2 - n_0^2 & -ic^2\mu_0\sigma_{xyz}q_z/\omega \\ ic^2\mu_0\sigma_{xyz}q_z/\omega & n^2 - n_0^2 \end{pmatrix}\boldsymbol{E} = 0, \tag{149}$$

We can then solve the refractive index

$$n_\pm = n_0 \pm \frac{c\mu_0}{2}\sigma_{xyz}. \tag{150}$$

The component of the optical conductivity σ_{xyz} can be certainly derived using the linear response theory [18, 19, 104]. However, here we will adopt the semiclassical theory at low frequency range (frequency is much smaller than the band gap), which is more intuitive and has clear physical interpretation for the result [18]. We will start with the current in Eq. (127). By plugging in the velocity in Eq. (128), we have

$$\boldsymbol{J}_1 = -e \int \frac{d\boldsymbol{k}}{8\pi^3}\left[\tilde{\boldsymbol{v}} + e\boldsymbol{E}\times\boldsymbol{\Omega}^{\mathrm{t}} + \frac{e}{\hbar}(\tilde{\boldsymbol{v}}\cdot\boldsymbol{\Omega}^{\mathrm{t}})\boldsymbol{B}\right]f. \tag{151}$$

Since $\boldsymbol{B}\propto q_z\boldsymbol{E}$, we should keep terms in the current that is either linear in the electric field or the magnetic field. The result reads

$$\boldsymbol{J}_1 = -e \int \frac{d\boldsymbol{k}}{8\pi^3}\left[\boldsymbol{v} - \boldsymbol{\partial}(\boldsymbol{B}\cdot\boldsymbol{m}) + e\boldsymbol{E}\times\boldsymbol{\Omega} + \frac{e}{\hbar}(\boldsymbol{v}\cdot\boldsymbol{\Omega})\boldsymbol{B}\right]f. \tag{152}$$

There is another contribution to the current due to the spatial average discussed in Sect. III E. The above current is only an accurate account for the first term in Eq. (87). The second term reads

$$\boldsymbol{J}_2 = \boldsymbol{\nabla}\times\int\frac{d\boldsymbol{k}}{(2\pi)^3}f\boldsymbol{m}. \tag{153}$$

Under the optical electric field, we can still use the Boltzmann equation to solve the distribution function, i.e.

$$\frac{\partial f}{\partial t} + \dot{\boldsymbol{k}}\cdot\frac{\partial f}{\partial\boldsymbol{k}} + \dot{\boldsymbol{r}}\cdot\frac{\partial f}{\partial\boldsymbol{r}} = -\frac{f - f_0}{\tau}. \tag{154}$$

Up to first order in the electric field, we can use the following ansatz for the distribution function

$$f = f_0 + f_1 e^{-i\omega t + i\boldsymbol{q}\cdot\boldsymbol{r}}. \tag{155}$$

The coefficient f_1 can then be solved through the Boltzmann equation. The result reads

$$\begin{aligned}f_1 &= -\frac{e\boldsymbol{E}\cdot\boldsymbol{v}}{i\omega - i\boldsymbol{q}\cdot\boldsymbol{v} - 1/\tau}f_0' \\ &= i\frac{e\boldsymbol{E}\cdot\boldsymbol{v}}{\omega}f_0' + i\frac{e\boldsymbol{E}\cdot\boldsymbol{v}}{\omega^2}\boldsymbol{q}\cdot\boldsymbol{v}f_0' + \cdots.\end{aligned} \tag{156}$$

In the second equality, we have absorbed the relaxation time into the imaginary part of the frequency, i.e. $\omega + i/\tau \to \omega$. The expansion is valid when the electron velocity $\boldsymbol{v}_0$ is much smaller than the speed of light.

By plugging the distribution function in Eq. (155) into the current in Eq. (152) and (153), we obtain

$$\begin{aligned}\boldsymbol{J} &= -e\int\frac{d\boldsymbol{k}}{8\pi^3}\left[i\boldsymbol{v}(\boldsymbol{q}\cdot\boldsymbol{v})\frac{e\boldsymbol{E}\cdot\boldsymbol{v}}{\omega}f_0' + \boldsymbol{v}(\boldsymbol{B}\cdot\boldsymbol{m})f_0' + \frac{e}{\hbar}(\boldsymbol{v}\cdot\boldsymbol{\Omega})\boldsymbol{B}f_0 + \boldsymbol{q}\times\boldsymbol{m}\frac{\boldsymbol{E}\cdot\boldsymbol{v}}{\omega}f_0'\right] \\ &= -\frac{e}{\omega}\int\frac{d\boldsymbol{k}}{8\pi^3}\left[\boldsymbol{v}(\boldsymbol{q}\times\boldsymbol{E}\cdot\boldsymbol{m})f_0' + \frac{e}{\hbar}(\boldsymbol{v}\cdot\boldsymbol{\Omega})\boldsymbol{q}\times\boldsymbol{E}f_0 + \boldsymbol{q}\times\boldsymbol{m}(\boldsymbol{E}\cdot\boldsymbol{v})f_0'\right].\end{aligned} \tag{157}$$

The equality holds as we only concern with the antisymmetric part of the optical conductivity and the first term in the original expression only contributes to the symmetric part. We also use the identity $\boldsymbol{B} = \boldsymbol{q}\times\boldsymbol{E}/\omega$. This current can also be derived using the linear response theory under appropriate limit [18, 19]. This current corresponds to the following optical conductivity

$$\sigma_{ijk} = -\frac{e}{\omega}\int\frac{d\boldsymbol{k}}{8\pi^3}\left[-\epsilon_{jk\ell}v_i m_\ell f_0' - \epsilon_{ijk}\frac{e}{\hbar}(\boldsymbol{v}\cdot\boldsymbol{\Omega})f_0 + \epsilon_{ik\ell}m_\ell v_j f_0'\right]. \tag{158}$$

It is evident that the above conductivity is antisymmetric about the indices i and j. The essential ingredient in the above expression takes the form of the dipole of Berry curvature (in the form of $\boldsymbol{v}\boldsymbol{\Omega}$) and magnetic moment (in the form of $\boldsymbol{v}\boldsymbol{m}$). Since the velocity, Berry curvature and magnetic moment all change sign under time reversal operation, and the velocity also change sign under inversion operation, the above current will require the system to break inversion symmetry instead of time reversal symmetry to become nonzero. Due to the antisymmetry between the indices i and j in σ_{ijk}, one can define a gyrotropic pseudotensor

$$g_{ij} = \frac{1}{2}\epsilon_{k\ell i}\sigma_{k\ell j} = \frac{e}{\omega}\int\frac{d\boldsymbol{k}}{8\pi^3}\left[\frac{e}{\hbar}(\boldsymbol{v}\cdot\boldsymbol{\Omega})f_0\delta_{ij} - \frac{1}{2}(m_i v_j + m_j v_i)f_0' + (\boldsymbol{m}\cdot\boldsymbol{v})\delta_{ij}f_0'\right]. \tag{159}$$

The trace of the gyrotropic tensor is a useful quantity as it corresponds to the conductivity that is proportional to ϵ_{ijk}, i.e. the optical current is always flowing in the direction of the magnetic field. Therefore, this is a robust contribution to the circular dichroism. Based on the above result, we have

$$\mathrm{Tr}g = \frac{e}{\omega} \int \frac{d\boldsymbol{k}}{8\pi^3} \left[3\frac{e}{\hbar}(\boldsymbol{v} \cdot \boldsymbol{\Omega})f_0 + 2(\boldsymbol{m} \cdot \boldsymbol{v})f_0' \right]. \qquad (160)$$

In the following, we consider an interesting scenario in the Weyl semimetal. The Weyl semimetal contains pairs of Weyl points near the Fermi level. Without loss of generality, we only consider one pair, described by the following Hamiltonian

$$\hat{H} = v(k_x \sigma_x \tau_z + k_y \sigma_y + k_z \sigma_z), \qquad (161)$$

where $\boldsymbol{\sigma}$ are pseudospin Pauli matrices and $\tau_z = \pm 1$ labels Weyl points with opposite chirality. We assume that the Fermi level falls in the conduction band. Then the Berry curvature and the magnetic moment reads

$$\boldsymbol{\Omega} = -\frac{\tau_z \boldsymbol{k}}{2k^3}, \quad \boldsymbol{m} = -\frac{\tau_z e v \boldsymbol{k}}{2\hbar k^2}. \qquad (162)$$

We then find that

$$\mathrm{Tr}g = -\sum_{\tau_z} \frac{e^2 \tau_z \mu(\tau_z)}{4\pi^2 \hbar \omega}, \qquad (163)$$

where $\mu(\tau_z)$ is the chemical potential for the Weyl node with the chirality τ_z, measured from the Weyl point.

This corresponds to the following current

$$\boldsymbol{J} = -\sum_{\tau_z} \frac{e^2 \tau_z \mu(\tau_z)}{12\pi^2 \hbar} \boldsymbol{B}. \qquad (164)$$

In equilibrium, the noncentrosymmetric Weyl semimetal has the two Weyl points with the same energy, as a result $\mu(+1) =$

$\mu(-1)$ and the above current vanishes. However, it can be nonzero in two cases: (i) the system can be driven out of equilibrium by applying the parallel electric and magnetic field simultaneously, which will create a finite difference between $\mu(+1)$ and $\mu(-1)$ due to the chiral anomaly [7, 8, 105–107]; (ii) in Weyl semimetals that simultaneously break the time reversal and inversion symmetry, the two Weyl points can sit on different energies, such that the same absolute value of the chemical potential can still result in different $\mu(+1)$ and $\mu(-1)$ when measured from Weyl points. However, in the latter case, one has to keep in mind this current only exists out of equilibrium with the optical fields applied.

Finally, we comment that the meaning of $\mathrm{Tr}g$ and hence σ_{ijk} can be viewed from a different aspect. The last Maxwell equation can be put in the following form:

$$\boldsymbol{\nabla} \times \boldsymbol{B} = \frac{1}{c^2}\frac{\partial \boldsymbol{E}}{\partial t} + \mu_0 \left(\frac{\partial \boldsymbol{P}}{\partial t} + \boldsymbol{\nabla} \times \boldsymbol{M} + \hat{e}_i \partial_j \frac{\partial Q_{ij}}{\partial t} \right), \qquad (165)$$

where Q_{ij} is the electric quadrupole moment. We can then identify the current as

$$\boldsymbol{J} = \frac{\partial \boldsymbol{P}}{\partial t} + \boldsymbol{\nabla} \times \boldsymbol{M} + \hat{e}_i \partial_j \frac{\partial Q_{ij}}{\partial t}. \qquad (166)$$

The material property $\boldsymbol{P}$, $\boldsymbol{M}$, and Q_{ij} can respond to the optical fields, through appropriate response functions. For the part responsible for the circular dichroism, we have

$$\begin{aligned} P_i &= \alpha_{ij}^{em} B_j, \\ M_j &= \alpha_{ij}^{me} E_i, \\ \dot{P}_i &= \beta_{ijk}(\partial_j E_k + \partial_k E_j), \\ \dot{Q}_{ij} &= \theta_{ijk} E_k. \end{aligned} \qquad (167)$$

As a result, we find that

$$J_i = -i\alpha_{ij}^{em}\epsilon_{jk\ell}q_k E_\ell + i\epsilon_{ijk}q_j\alpha_{k\ell}^{me}E_\ell + i\beta_{ijk}(q_j E_k + q_k E_j) + iq_j\theta_{ijk}E_k. \qquad (168)$$

For the trace of g, the last two terms do not contribute due to the anti-symmetrization of indices. We finally find that

$$\mathrm{Tr}g = 2i\mathrm{Tr}(\alpha_{ij}^{em} - \alpha_{ij}^{me}). \qquad (169)$$

This suggests that the circular dichroism is a measure of the difference between different types of dynamical magnetoelectric couplings [104].

E. Spatially dispersive phenomenon: nonreciprocal directional dichroism in multiferroics

In multiferroics where both the time-reversal symmetry and the inversion symmetry are broken, there is a unique spatially dispersive phenomenon known as nonreciprocal directional dichroism (NDD) [108]. It refers to different refractive indices for counter-propagating lights. Since it requires

both broken time-reversal and inversion symmetry, which renders the co-existence of the polarization and magnetization, NDD can probe the dynamical coupling between electricity and magnetism in multiferroics [109–117]. Previously, we have shown that the circular dichroism in noncentrosymmetric materials is determined by the dipole of Berry curvature and magnetic moment. It is interesting to explore the geometrical quantities that determine NDD.

We begin with a brief phenomenological derivation of NDD in the electromagnetic theory. We still consider the wave equation for the electric field

$$\boldsymbol{\nabla} \times (\boldsymbol{\nabla} \times \boldsymbol{E}) = -\mu_0 \frac{\partial \boldsymbol{J}}{\partial t} - \frac{1}{c^2}\frac{\partial^2 \boldsymbol{E}}{\partial t^2}. \qquad (170)$$

We will assume that a monochromatic light is involved, which is linearly polarized along the x-direction and propagating along the z-direction.
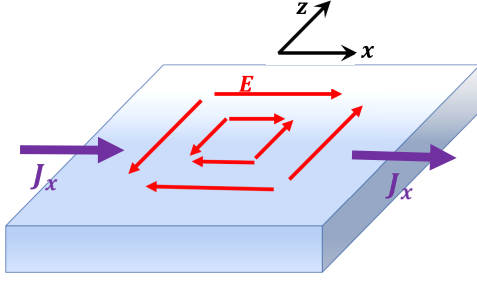
FIG. 7. The electric quadrupolar current. The electric field in red arrows has the quadrupolar profile. The resulting quadrupolar current is in purple arrows. From Ref. [20].

Without loss of generality, we can exert the following symmetry constraints to simplify the result. First, we can assume the mirror-z symmetry is broken but mirror-x symmetry is present. This forbids the existence of σ_{xy}. We further assume that there is rotational symmetry about the z-axis, so that $\sigma_{xxz} = \sigma_{yyz}$. Then the above symmetry assumptions yield the following equation for the refractive index: $n_z^2 = n_0^2 + ic\mu_0 n_z \sigma_{xxz}$. The subscript z in n_z is added to distinguish the direction of propagation. The solution is

$$n_z = ic\mu_0\sigma_{xxz}/2 + \sqrt{n_0^2 - c^2\mu_0^2\sigma_{xxz}^2/4}\,. \qquad (171)$$

By reversing the propagation direction, the first term in Eq. (171) flips sign and we can then solve n_{-z}. The difference is

$$\Delta n = n_z - n_{-z} = ic\mu_0\sigma_{xxz}\,. \qquad (172)$$

We will consider the static transport part in σ_{xxz}. By definition, it corresponds to the following current $J_x \propto q_z E_x \propto \partial_z E_x$, i.e. the current driven by a spatially varying but static electric field in metals. In the static case, $\nabla \times \boldsymbol{E} = -\partial \boldsymbol{B}/\partial t = 0$, the electric field must have a quadrupolar profile as shown in Fig. 7. This can be depicted by the semiclassical transport theory in Sect. III C. Moreover, the band geometry will enter naturally in the semiclassical equations of motion. The equations of motion for Bloch electrons in the m-th band under slowly varying electric field read

$$\dot{\boldsymbol{r}} = \frac{1}{\hbar}\partial_k\tilde{\varepsilon}_m - \dot{\boldsymbol{k}}\times\boldsymbol{\Omega}_m - \boldsymbol{\Omega}_{\boldsymbol{kr},m}\cdot\dot{\boldsymbol{r}}\,, \qquad (173)$$

$$\hbar\dot{\boldsymbol{k}} = -e\boldsymbol{E} + \boldsymbol{\Omega}_{\boldsymbol{rk},m}\cdot\hbar\dot{\boldsymbol{k}}\,, \qquad (174)$$

where $\boldsymbol{\Omega}_m = -2\mathrm{Im}\langle\partial_k u_m|\times|\partial_k u_m\rangle$ is the momentum-space Berry curvature, and $(\Omega_{\boldsymbol{kr},m})_{ij} = -2\mathrm{Im}\langle\partial_{k_i} u_m|\partial_{r_j} u_m\rangle$ is the mixed Berry curvature with $(\Omega_{\boldsymbol{kr},m})_{ij} = -(\Omega_{\boldsymbol{rk},m})_{ji}$. $\boldsymbol{\Omega}_{\boldsymbol{kr},m}$ arises as the inhomogeneous electric field modifies the Bloch wave function.

We now calculate the modified band energy, i.e. $\tilde{\varepsilon}_m = \varepsilon_m + \delta\varepsilon$. The electrostatic potential $\phi(\boldsymbol{r})$ can be expanded around the center of mass position of the wave packet

$$\phi(\boldsymbol{r}) = \phi(\boldsymbol{r}_c) + e\boldsymbol{E}(\boldsymbol{r}_c)\cdot(\boldsymbol{r}-\boldsymbol{r}_c)$$
$$+ \frac{1}{2}e[\partial_i E_j(\boldsymbol{r}_c)](\boldsymbol{r}-\boldsymbol{r}_c)_i(\boldsymbol{r}-\boldsymbol{r}_c)_j + \cdots\,, \qquad (175)$$

where i and j label the spatial directions. The energy correction can be obtained by calculating the expectation value of the third term in Eq. (175) under the wave packet state:

$$\delta\varepsilon = \frac{e}{2}\partial_i E_j\int d\boldsymbol{k}d\boldsymbol{k}'\,C_m^\star(\boldsymbol{k}')C_m(\boldsymbol{k})\langle u_m(\boldsymbol{k}')|(i\partial_{k_i'}-r_{ci})e^{-i\boldsymbol{k}'\cdot\boldsymbol{r}}(-i\partial_{k_j}-r_{cj})e^{i\boldsymbol{k}\cdot\boldsymbol{r}}|u_m\rangle$$

$$= \frac{e}{2}\partial_i E_j\int d\boldsymbol{k}d\boldsymbol{k}'\,(-i\partial_{k_i'}-r_{ci})C_m^\star(\boldsymbol{k}')(i\partial_{k_j}-r_{cj})C_m(\boldsymbol{k})\langle u_m(\boldsymbol{k}')|e^{-i\boldsymbol{k}'\cdot\boldsymbol{r}}e^{i\boldsymbol{k}\cdot\boldsymbol{r}}|u_m\rangle$$

$$+ \frac{e}{2}\partial_i E_j\int d\boldsymbol{k}d\boldsymbol{k}'\,C_m^\star(\boldsymbol{k}')(-i\partial_{k_j}-r_{cj})C_m(\boldsymbol{k})(-i)\langle\partial_{k_i'}u_m(\boldsymbol{k}')|e^{-i\boldsymbol{k}'\cdot\boldsymbol{r}}e^{i\boldsymbol{k}\cdot\boldsymbol{r}}|u_m\rangle$$

$$+ \frac{e}{2}\partial_i E_j\int d\boldsymbol{k}d\boldsymbol{k}'\,(-i\partial_{k_i'}-r_{ci})C_m^\star(\boldsymbol{k}')C_m(\boldsymbol{k})\langle u_m(\boldsymbol{k}')|e^{-i\boldsymbol{k}'\cdot\boldsymbol{r}}e^{i\boldsymbol{k}\cdot\boldsymbol{r}}i|\partial_{k_j}u_m\rangle$$

$$+ \frac{e}{2}\partial_i E_j\int d\boldsymbol{k}d\boldsymbol{k}'\,C_m^\star(\boldsymbol{k}')C_m(\boldsymbol{k})(-i)\langle\partial_{k_i'}u_m(\boldsymbol{k}')|e^{-i\boldsymbol{k}'\cdot\boldsymbol{r}}e^{i\boldsymbol{k}\cdot\boldsymbol{r}}i|\partial_{k_j}u_m\rangle$$

$$= \frac{e}{2}\partial_i E_j\int d\boldsymbol{k}\,C_m^\star(\boldsymbol{k})C_m(\boldsymbol{k})\sum_n^{n\neq m}(-i)\langle\partial_{k_i}u_m|u_n\rangle\langle u_n|i|\partial_{k_j}u_m\rangle$$

$$= \frac{e}{2}\partial_i E_j g_{ij,m}\,. \qquad (176)$$

Here $g_{ij,m}$ is the quantum metric tensor for band m defined in Eq. (24). As the quantum metric couples to $\partial_i E_j$ in the energy, it can be viewed as the electric quadrupole of Bloch states, similar to the definition of the electric quadrupole in the electromagnetic theory.

Based on the discussion in Sect. III E, the local current in the semiclassical theory in inhomogeneous systems are given by

$$\boldsymbol{J}_{\mathrm{tr}} = -e\sum_m\int\frac{d\boldsymbol{k}}{8\pi^3}D\dot{\boldsymbol{r}}f_m$$

$$-\nabla_r \times \frac{e}{\hbar} \sum_m \int \frac{d\mathbf{k}}{8\pi^3} k_B T \mathbf{\Omega}_m \log\left(1 + e^{\frac{\mu - \varepsilon_m}{k_B T}}\right), \qquad (177)$$

where f_m is the Fermi function, and $D = 1 + \mathrm{Tr}\Omega_m^{kr}$ is modified density of states. Here we have discounted the magnetization current $\nabla \times \mathbf{M}$.

To evaluate Eq. (177) at the order ∂E, we need carefully evaluate all components in the correct order. The velocity should contain the band velocity from $\delta\varepsilon$ and the anomalous velocity due to Ω_m^{kr}. The former one is easy to implement. For the latter one, we have $\Omega_m^{kr} = -\partial_r \mathcal{A}^k$. To evaluate this quantity at the order of ∂E, we need to use the field correction to the Berry connection, i.e. the positional shift given in Eq. (107). The result is

$$(\Omega_m^{kr})_{ij} = -2e\partial_j E_\ell \mathrm{Re} \sum_n^{n \neq m} \frac{(\mathcal{A}_i)_{mn}(\mathcal{A}_\ell)_{nm}}{\varepsilon_m - \varepsilon_n}. \qquad (178)$$

We also need the response of the Berry curvature to the electric field, i.e. the curl of the positional shift given in Eq. (107)

$$\delta\Omega_m' = \nabla \times \mathcal{A}'$$
$$= 2e\nabla_k \times \mathrm{Re} \sum_{n \neq m} \frac{\mathcal{A}_{mn}(\mathcal{A}_{nm} \cdot \mathbf{E})}{\omega_{mn}}. \qquad (179)$$

where $\omega_{mn} = \varepsilon_m - \varepsilon_n$.

These are the all ingredients required to obtain the current at the order of ∂E. The derivation can be done as follows

$$
\begin{aligned}
-e\sum_m \int \frac{d\mathbf{k}}{8\pi^3} D\dot{r}_i f_m =& 2\frac{e^2}{\hbar}\partial_j E_\ell \sum_m \int \frac{d\mathbf{k}}{8\pi^3} f_m \sum_n^{n \neq m} \mathrm{Re} \frac{v_{i,m}(\mathcal{A}_j)_{mn}(\mathcal{A}_\ell)_{nm} - v_{j,m}(\mathcal{A}_i)_{mn}(\mathcal{A}_\ell)_{nm}}{\varepsilon_m - \varepsilon_n} \\
&- \frac{e^2}{2\hbar}\sum_m \partial_j E_k \int \frac{d\mathbf{k}}{8\pi^3} f_m \partial_i g_{jk,m}, \\
=& 2\frac{e^2}{\hbar}\partial_z E_x \sum_m \int \frac{d\mathbf{k}}{8\pi^3} \sum_n^{n \neq m} \mathrm{Re} \frac{v_{x,m}(\mathcal{A}_z)_{mn}(\mathcal{A}_x)_{nm} - v_{z,m}(\mathcal{A}_x)_{mn}(\mathcal{A}_x)_{nm}}{\varepsilon_m - \varepsilon_n} f_m \\
&+ \frac{e^2}{\hbar}\partial_z E_x \sum_m \int \frac{d\mathbf{k}}{8\pi^3} f_m' v_{x,m} g_{xz,m}. \qquad (180)
\end{aligned}
$$

Since our purpose is to obtain the *xxz* component of the conductivity, we assign ijk to *xxz* and *xzx* and use the fact that $\partial_z E_x = \partial_x E_z$ for static electric fields. Then we have

$$
\begin{aligned}
\frac{e}{\hbar}\left(\nabla_r \times \sum_m \int \frac{d\mathbf{k}}{8\pi^3} \mathcal{F}_m \delta\Omega_m\right)_x &= -2\frac{e^2}{\hbar}\partial_z E_x \sum_m \int \frac{d\mathbf{k}}{8\pi^3} \mathcal{F}_m \left(\nabla_k \times \mathrm{Re} \sum_n^{n \neq m} \frac{\mathcal{A}_{mn}(\mathcal{A}_x)_{nm}}{\varepsilon_m - \varepsilon_n}\right)_y \\
&= -2\frac{e^2}{\hbar}\partial_z E_x \sum_m \int \frac{d\mathbf{k}}{8\pi^3} f_m \mathrm{Re} \sum_n^{n \neq m} \frac{v_{x,m}(\mathcal{A}_z)_{mn}(\mathcal{A}_x)_{nm} - v_{z,m}(\mathcal{A}_x)_{mn}(\mathcal{A}_x)_{mn}}{\varepsilon_m - \varepsilon_n},
\end{aligned}
\qquad (181)
$$

where $\mathcal{F}_m = -k_B T \log[1 + \exp((\mu - \varepsilon_m)/k_B T)]$ and we have used integration by parts in the second line.

We find that Eq. (181) cancels the first term in Eq. (180). Therefore, only the contribution from $\delta\varepsilon$ remains in transport [20, 118]:

$$J_x^{\mathrm{tr}} = -2e \int \frac{d\mathbf{k}}{8\pi^3} f_m \partial_{k_x} \delta\varepsilon = \gamma_{xxz}\partial_z E_x, \qquad (182)$$

where

$$\gamma_{jik} = \frac{e^2}{\hbar}\sum_m \int \frac{d\mathbf{k}}{8\pi^3} G_{ijk,m} f_m', \qquad (183)$$

Here $G_{ijk,m} = v_{i,m} g_{jk,m}$ is the quantum metric dipole, defined similarly to the Berry curvature dipole [9]. The factor 2 in

Eq. (182) is due to $\partial_z E_x = \partial_x E_z$. As discussed previously, the quantum metric is the Bloch state quadrupole. Therefore, $G_{ijk,m}$ can also be viewed as the momentum space dipole of the electric quadrupole.

Equation (182) is independent of the transport relaxation time, i.e. it is an intrinsic current. The quantum metric does not change under either the time reversal or inversion operation but the velocity will change sign in both cases. As a result, the quantum metric dipole flips sign under time reversal and inversion operation. Moreover, the quantum metric dipole $G_{xxz,m}$ contains one derivative along z direction, and hence it flips sign under the mirror-z operation. Therefore, the current in Eq. (182) is only nonzero only when the time reversal symmetry, the inversion symmetry, and the mirror-z symmetry are
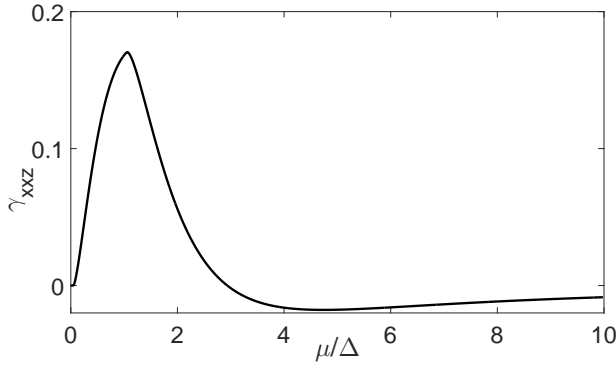
FIG. 8. The electric quadrupolar current in the low-energy model. The parameters are chosen as follows: $\Delta_1 = \Delta$, $\Delta_2 = 2\Delta$, $v'_1 = -v'_2 = v'$, $\frac{1}{2}m(v')^2 = 0.95\Delta$. The system has a small global band gap 0.1Δ. γ_{xxz} is in units of $e^2/(16\pi^2\hbar)$. From Ref. [20].

all broken. Due to the appearance of f'_m, this current is also a Fermi surface property and will be important in metals and semiconductors. Since this current is proportional to the variation of the electric field not the electric field itself, it can exist even when there is no net voltage applied across the sample, i.e. no net electric field.

As a concrete example, we consider the following Hamilontian

$$\hat{H} = v'_i k_z + v\tau_i k_x \sigma_x + v k_y \sigma_y + \left(\Delta_i + \frac{k_z^2}{2m_i}\right)\sigma_z, \qquad (184)$$

where i is the valley index, $\tau_i = \pm 1$, $2\Delta_i$ is the band gap, v is the in-plane Fermi velocity, and $m_i > 0$ is the effective mass along the z-direction. The first term introduces a tilting and breaks the mirror-z symmetry for each valley. This model can be realized by stacking certain 2D magnets [119, 120] along the z-direction and make the stacking order break the mirror-z symmetry.

In Fig. 8, we can see the behavior of the relevant conductivity γ_{xxz} as a function of the chemical potential at zero temperature. When μ lies in the global band gap, $\gamma_{xxz} = 0$ as there is no Fermi surface. As μ crosses the conduction band of one valley but is still inside the gap of the other valley, γ_{xxz} increases because of the increasing density of states. When μ further increases to intersect the conduction bands of both valleys, the other valley will also contribute but with opposite signs. As a result γ_{xxz} begins to decrease. When μ is far away from the band edge, γ_{xxz} approaches 0, because the quantum metric dipole decays faster than the increasing density of states.

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