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Linear and Nonlinear Electromagnetic Responses in Topological Semimetals

by

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Professor Joel E. Moore, Chair
Professor Dung-Hai Lee
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Abstract

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The topological consequences of time reversal symmetry breaking in two dimensional electronic systems have been a focus of interest since the discovery of the quantum Hall effects. Similarly interesting phenomena arise from breaking inversion symmetry in three dimensional systems. For example, in Dirac and Weyl semimetals the inversion symmetry breaking allows for non-trivial topological states that contain symmetry-protected pairs of chiral gapless fermions. This thesis presents our work on the linear and nonlinear electromagnetic responses in topological semimetals using both a semiclassical Boltzmann equation approach and a full quantum mechanical approach. In the linear response, we find a "gyrotropic magnetic effect" (GME) where the current density j^B in a clean metal is induced by a slowly-varying magnetic field. It is shown that the experimental implications and microscopic origin of GME are both very different from the chiral magnetic effect (CME). We develop a systematic way to study general nonlinear electromagnetic responses in the low-frequency limit using a Floquet approach and we use it to study the circular photogalvanic effect (CPGE) and second-harmonic generation (SHG). Moreover, we derive a semiclassical formula for magnetoresistance in the weak field regime, which includes both the Berry curvature and the orbital magnetic moment. Our semiclassical result may explain the recent experimental observations on topological semimetals. In the end, we present our work on the Hall conductivity of insulators in a static inhomogeneous electric field and we discuss its relation to Hall viscosity.

To my grandfather,

Contents

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Chapter 1

Introduction

1.1 Background and motivation

According to the electronic properties, solid materials are divided into insulators, semiconductors, semimetals and metals (conductors). During the past decade, topology has become an important role in defining new phases of the matter and a new phase of matter called topological insulators has started to spurring researchers' interest. Different from conventional insulators, topological insulators have gapless surface states and quantized electronic response functions. These properties are protected by topology, meaning that under a smooth deformation (with the insulating gap open) of the Hamiltonian without breaking certain symmetries one can not turn a topological class of insulators into another class, for example, turning the topological insulators into conventional trivial insulators.

Similarly interesting phenomena have been found beyond insulating phases recently. An example is the Weyl semimetal in three dimensional space. In the band structure of a Weyl semimetal the conduction bands and valence bands intersect at isolated points. Those gapless points named as Weyl nodes are topologically protected in the sense that one can not transform a Weyl semimetal into an insulator by small perturbations when charge conservation and transnational invariance is preserved. Weyl semimetals also have protected surface states as in topological insulators.

The protection can be seen more clearly if one looks close to the Hamiltonian near a Weyl node at momentum $\mathbf{k}^*$:

$$H = \sum_{ij} \hbar \sigma_i b_{ij} (k_j - k_j^*) \tag{1.1}$$

where σ_i are Pauli matrices in the basis of the two bands involved and b_{ij} are some real parameters. As we can see, all Pauli matrices have been used up in this Hamiltonian, so one can not gap out the spectrum by adding anticommuting matrices. Using this Hamiltonian one can also define the chirality of the Weyl node by $\chi = \operatorname{sign} \det b_{ij}$. The chirality is the magnetic charge of a Weyl node: if one integrate the Berry curvature (It is like the magnetic field and we will talk more about it in the next chapter.) over a closed surface enclosing

the Weyl node the value will be $2\pi\chi$.

Recently, the condensed matter community has been actively studying the unusual transport and optical phenomena in Weyl semimetals. One important example is the chiral magnetic effect (CME) where Weyl nodes with different chemical potentials and a static magnetic field give rise to an electric current parallel to the magnetic field. Another example is the negative magnetoresistance where an applied static magnetic field leads to a large magnetoresistance with unusual anisotropy. However, these phenomena in Weyl semimetals has not been fully understood. In this thesis, we will try addressing the following concerns: (i) Recently, there is a claim stating that, in Weyl semimetals, a static magnetic field by itself is sufficient to drive a dissipationless current. Such an effect has been characterized as a type of CME. However, even the existence of this type of CME is questionable since it violates the Bloch theorem, which forbids the macroscopic currents in a bulk system in equilibrium. While this has been actively debated, no thorough study has been carried out. (ii) Recent research on the negative magnetoresistance in weak magnetic field using semiclassical approach has considered only chiral anomaly (or Berry curvature). However, in a complete analysis, another important component—orbital magnetic moment [67]—is indispensable. (iii) The semiclassical approach (SCA) has been used to study the nonlinear optical responses in metals. The SCA conserves the properly defined volume in phase space and gives an intuitive approach to many observable properties of metals. However, the SCA can make erroneous predictions if used on nonlinear optical responses in metals. A full quantum theory approach is necessary to confirm the validity of the results derived by the SCA.

In addition to studying the topological properties of the topological phases, an active area of the recent research has been studying the geometrical properties of the topological phase, such as the Hall viscosity. One question remained for the Hall viscosity is how to measure it in experiment. Recent work[66, 15] shows that the Hall conductivity in an inhomogenous electric field receives a contribution from the Hall viscosity. Therefore, the Hall viscosity can be determined by measuring the the Hall conductivity. At the end of this thesis we will present our work on the study of Hall conductivity in a inhomogenous electric field.

1.2 Overview

In this section we give an overview of the following chapters in this thesis.

Chapter 2 gives a brief review of the Berry phase, Berry curvature, quantum geometric tensor, and quantum metric tensor. We give a short introduction of the semiclassical theory, a tool we will use in the following chapters. At the end of this chapter we give an example of using semiclassical equations to solve a transport problem in metals.

In Chapter 3, we study the linear electromagnetic response in metals using semiclassical approach and a full quantum derivation. We find a "gyrotropic magnetic effect" (GME) where the current density j^B in a clean metal is induced by a slowly-varying magnetic field. We make a distinction between the chiral magnetic effect (CME) and GME. and by doing this we confirm that there can not be a dissipationless current in equilibrium. In the end

we work on a simple two band model of the Weyl semimetals and discuss the realization of GME as the low frequency limit of natural optical activity (NOA) on mirror-free Weyl semimetals.

In the supplemental material, we give the technical details of using Kubo formula to calculate the linear-response of natural gyrotropy in metals at low frequencies. We also give the derivation of a formula for the equilibrium optical activity coefficient of an isotropic Weyl semimetal and the derivation of a formula for the nonequilibrium optical gyrotropy of a Weyl semimetal due to the chiral anomaly. In addition to the technical details, we discuss the reciprocity relation for a metal. The Onsager reciprocity principle requires that in T-invariant materials there is no optical rotation in reflection (Kerr effect). We show how this principle is reflected in our semiclassical results.

In Chapter 4, we study the angle-dependent magnetoresistance in topological semimetals using a semiclassical approach. We apply our semiclassical result on the Hamiltonian for a Weyl semimetal and show that our result captures the directional anisotropy of linear conductivity under a magnetic field, which may explain the low-field regime of recent experiments. In this chapter we also study the nonlinear optical responses using a Floquet approach. Our approach reproduces the semiclassical formula for the circular photogalvanic effect (CPGE) and provides a systematic way to study general nonlinear electromagnetic responses in the low-frequency limit.

Chapter 5 studies the Hall conductivity of insulators in an inhomogeneous electric field to the second order of the wave number q using both the Kubo formula and the semiclassical approach. We show that in a special case when the system have only two bands and when the bands are all flat the q^2 term of the Hall conductivity is fully determined by an interplay of the Berry curvature and the quantum metric tensor. In the end we do numerical studies on the Chern insulators and discuss the relation between Hall conductivity and Hall viscosity.

1.3 List of publications

We list the publications for this thesis:

1. Semiclassical theory of nonlinear magneto-optical responses with applications to topological Dirac/Weyl semimetals, Takahiro Morimoto, Shudan Zhong, Joseph Orenstein and Joel E. Moore, *Phys. Rev. B*, 94, 245121 (2016).

2. Gyrotropic magnetic effect and the orbital moment on the Fermi surface,Shudan Zhong, Joel E. Moore, and Ivo Souza, *Physical Review Letter*, 116, 077201 (2016).

3. Optical Gyrotropy from Axion Electrodynamics in Momentum Space, Shudan Zhong, Joseph Orenstein and Joel E. Moore, *Physical Review Letter*, 115, 117403 (2015).

Chapter 2

Preliminaries

In this chapter, We will briefly review some basic techniques and concepts used in this thesis.

2.1 Berry phase

In 1984, Berry brings a geometrical phase factor to physicists' attention in his famous paper [11]. In the paper, the geometrical phase factor is shown exists in the adiabatic evolution of a quantum system in an eigenstate when the path of the evolution makes up a circuit in the Hamiltonian's parameter space. The phase is also called Berry phase and it origins from the nontrivial topological property of the parameter space. One example is the Aharonov-Bohm(AB) effect where the Berry phase is given by a line integral of the vector potential $\mathbf{A}$ along a closed loop,

$$\phi = \frac{q}{\hbar} \oint_C \mathbf{A}(\boldsymbol{R}) \cdot d\boldsymbol{R}\,. \tag{2.1}$$

This phase is gauge invariant and hence physical. One can check it by turning the line integral into a surface integral of the magnetic field $\mathbf{B}$. Indeed, Berry phase can generally be written as an integral of a field on the parameter space. The field is the Berry curvature. Berry curvature is also gauge invariant and it is the antisymmetric part of a gauge invariant quantum geometric tensor [12]

$$T_{ij} = \langle \partial_i n | (1 - |n\rangle\langle n|) | \partial_j n \rangle \tag{2.2}$$

where $|n\rangle$ denotes an eigenstate. The symmetric part of this tensor is called the quantum metric tensor. It provides a natural means to measure the distances between states.

2.2 Semiclassical theory

2.2.1 Bloch waves

In a crystal, atoms form a lattice which has discrete translational symmetry. The electrons in a lattice will feel a periodic potential

$$V(\mathbf{r}+\mathbf{R}) = V(\mathbf{r}) \tag{2.3}$$

where $\mathbf{R}$ is a lattice vector. With the assumption that the electrons we consider are free, meaning that they do not interact with each other (the interactions between electrons is assumed to be represented by an effective one electron potential that is included in V), the eigenstates of the Hamiltonian for this system will be Bloch waves according to Bloch's theorem

$$H\Psi_{n\mathbf{k}}(\mathbf{r}) = \epsilon_{n\mathbf{k}}\Psi_{n\mathbf{k}}(\mathbf{r}), \quad \Psi_{n\mathbf{k}}(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}}u_{n\mathbf{k}}(\mathbf{r})\,. \tag{2.4}$$

The $u_{n\mathbf{k}}(\mathbf{r})$ is periodic

$$u_{n\mathbf{k}}(\mathbf{r}+\mathbf{R}) = u_{n\mathbf{k}}(\mathbf{r})\,. \tag{2.5}$$

One can check that $\Psi_{n\mathbf{k}}$ is also the eigenstate of the translational operator T, which can be inferred from group theory

$$T\Psi_{n\mathbf{k}}(\mathbf{r}+\mathbf{R}) = \Psi_{n\mathbf{k}}(\mathbf{r}+\mathbf{R}) = e^{i\mathbf{k}\cdot(\mathbf{r}+\mathbf{R})}u_{n\mathbf{k}}(\mathbf{r}+\mathbf{R}) = e^{i\mathbf{k}\cdot\mathbf{R}}\Psi_{n\mathbf{k}}(\mathbf{r})\,. \tag{2.6}$$

As a consequence, the $\mathbf{k}$ is called the crystal momentum.

The set of all vectors $\mathbf{G}$ that satisfied

$$\mathbf{G}\cdot\mathbf{R} = 2\pi N, \quad N\in\mathbb{Z} \tag{2.7}$$

gives the reciprocal lattice. It can be shown that $\mathbf{k}$ is periodic: $\mathbf{k}+\mathbf{G}$ is the same as $\mathbf{k}$. Therefore, we usually limit the values of k to be in any primitive unit cell of the reciprocal lattice. There is one specific choice of the primitive unit cell called the Brillouin zone.

2.2.2 Semiclassical equations

In semiclassical theory, an electron is modeled as a wave packet. The wave packet is constructed from Bloch waves

$$|\Psi(\mathbf{r}_c,\mathbf{k}_c)\rangle = \int [d\mathbf{k}]\, a(\mathbf{k})\Psi_{n\mathbf{k}}(\mathbf{r},t)\,. \tag{2.8}$$

It is centered at $\mathbf{r}_c$ in real space and $\mathbf{k}_c$ in momentum space. $a(\mathbf{k})$ is arbitrary function of $\mathbf{k}$ as long as the spread of the wave packet in $\mathbf{k}$ space is narrow compared with the Brillouin zone.

The semiclassical equations of motion for the centers r_c and k_c of an electron wave packet are [67]

$$\dot{\boldsymbol{r}} = \frac{1}{\hbar}\boldsymbol{\nabla}_{\boldsymbol{k}}\epsilon_{\boldsymbol{k}} - \dot{\boldsymbol{k}} \times \boldsymbol{\Omega}, \tag{2.9}$$

$$\dot{\boldsymbol{k}} = -e\boldsymbol{E} - e\dot{\boldsymbol{r}} \times \boldsymbol{B}. \tag{2.10}$$

where

$$\boldsymbol{\Omega} = -\mathrm{Im}[\langle \nabla_{\boldsymbol{k}} u_{\boldsymbol{k}}| \times |\nabla_{\boldsymbol{k}} u_{\boldsymbol{k}}\rangle] \tag{2.11}$$

is the Berry curvature and the electron charge is (−e). We omit the subscript c for the wave packet centers and n for the Bloch waves.

From the symmetry of these equations between position and momentum, it is clear that $\boldsymbol{\Omega}(\mathbf{k})$ can be viewed as an effective magnetic field in momentum space and the magnetic field is the Berry curvature in real space.

One can see this symmetry more clearly with the Berry connection defined as

$$\mathcal{A}(\mathbf{k}) = i\langle u_{\mathbf{k}}|\boldsymbol{\nabla}_{\mathbf{k}}|u_{\mathbf{k}}\rangle\,. \tag{2.12}$$

The Berry connection can be viewed as the vector potential $\mathbf{A}$ in momentum space. The Berry curvature will be given by

$$\boldsymbol{\Omega}(\mathbf{k}) = \boldsymbol{\nabla}_{\mathbf{k}} \times \mathcal{A}(\mathbf{k})\,. \tag{2.13}$$

We know that in real space the Schrodinger equation

$$i\partial_t \Psi(\mathbf{r},t) = H(\mathbf{r})\Psi(\mathbf{r},t) = -\frac{\nabla^2}{2m}\Psi(\mathbf{r},t) + V(\mathbf{r})\Psi(\mathbf{r},t) \tag{2.14}$$

is invariant under a global phase shift

$$\Psi(r,t) \to e^{i\phi}\Psi(r,t) \tag{2.15}$$

since ϕ is a constant. In addition, the Schrodinger equation is also invariant under local phase shift if it is for a charge particle. If we do a local phase shift to the wave function of a charge particle

$$\Psi(r,t) \to e^{i\phi(\mathbf{r},t)}\Psi(r,t) \tag{2.16}$$

the Schrodinger equation will be invariant if we also do a gauge transformation of the potential φ and $\mathbf{A}$

$$\begin{aligned} \varphi' &= \varphi - \frac{1}{q}\partial_t\phi(\mathbf{r},t) \\ \mathbf{A}' &= \mathbf{A} + \frac{1}{q}\nabla\phi(\mathbf{r},t)\,. \end{aligned} \tag{2.17}$$

One can check that

$$(i\partial_t - q\varphi')e^{i\phi(\mathbf{r},t)}\Psi(r,t) = e^{i\phi(\mathbf{r},t)}(i\partial_t - q\varphi' - \partial_t\phi(\mathbf{r},t))\Psi(r,t) \tag{2.18}$$

$$[\frac{1}{2m}(-i\boldsymbol{\nabla} - q\mathbf{A}')^2 + V]e^{i\phi(\mathbf{r},t)}\Psi(r,t) = e^{i\phi(\mathbf{r},t)}[\frac{1}{2m}(-i\boldsymbol{\nabla} - q\mathbf{A}' + \nabla\phi(\mathbf{r},t))^2 + V]\Psi(r,t)\,. \tag{2.19}$$

With Eq. (2.17) we have

$$(i\partial_t - q\varphi')e^{i\phi(\mathbf{r},t)}\Psi(r,t) = [\frac{1}{2m}(-i\boldsymbol{\nabla} - q\mathbf{A}')^2 + V]e^{i\phi(\mathbf{r},t)}\Psi(r,t) \tag{2.20}$$

indicating that the Schrodinger equation is invariant. Using Eq. (2.12) a similar transformation can be done for a Bloch wave

$$\begin{aligned} |u_\mathbf{k}\rangle &\to e^{i\phi(\mathbf{k})}|u_\mathbf{k}\rangle \\ \mathcal{A} &\to \mathcal{A} - \nabla_k\phi(\mathbf{k})\,. \end{aligned} \tag{2.21}$$

The minus sign can be removed by redefining $\mathcal{A}$ but we would like to keep the current definition for it is used in the following chapters. It can be shown that the Bloch wave with a local phase shift is still the solution of the Hamiltonian. The symmetry between position and momentum in semiclassical equations can then be seen more clearly from

$$\begin{aligned} \dot{\mathbf{r}} &= \nabla_\mathbf{k}\epsilon_\mathbf{k} - \dot{\mathbf{k}} \times (\nabla_\mathbf{k} \times \mathcal{A}) \\ \dot{\mathbf{k}} &= e\nabla_\mathbf{r}\phi - e\dot{\mathbf{r}} \times (\nabla_\mathbf{r} \times \mathbf{A})\,. \end{aligned} \tag{2.22}$$

There are two more important things one should pay attention to when using the semiclassical equations. One is that the energy $\epsilon_\mathbf{k}$ in Eq. (2.10) has a contribution from the orbital magnetic moment [67],

$$\epsilon_\mathbf{k} = \epsilon^0_\mathbf{k} - \boldsymbol{m}_k \cdot \mathbf{B} \tag{2.23}$$

where $H_{\boldsymbol{k}}|u_{\boldsymbol{k}}\rangle = \epsilon^0_{\boldsymbol{k}}|u_{\boldsymbol{k}}\rangle$ with $B = 0$ and the orbital magnetic moment is given by

$$\boldsymbol{m}_{\boldsymbol{k}} = -\frac{e}{2}\mathrm{Im}[\langle\nabla_{\boldsymbol{k}}u_{\boldsymbol{k}}| \times (H_{\boldsymbol{k}} - \epsilon^0_{\boldsymbol{k}})|\nabla_{\boldsymbol{k}}u_{\boldsymbol{k}}\rangle]. \tag{2.24}$$

The orbital magnetic moment represents the self-rotation of a wave packet.

The other thing is that one should use a modified phase-space density which is induced by an external magnetic field [77]

$$D = 1 + e\mathbf{B} \cdot \boldsymbol{\Omega} \tag{2.25}$$

in order to have the phase space volume conserved.

2.2.3 Example: the response to a static magnetic field

The semiclassical equations with the Boltzmann equation have been successfully used to understand various transport phenomena of solids, such as Haas-van Alphen oscillation, anomalous Hall effect, etc. In this section, we will give an example of using semiclassical equations to study the response to a static magnetic field.

In the presence of a static magnetic field, the electrons are in an equilibrium state with the distribution function $f(\epsilon_{\mathbf{k}})$. We solve $\dot{\mathbf{r}}$ using Eq. (2.10):

$$\dot{\mathbf{r}} = \frac{1}{D}[\nabla_{\mathbf{k}}\epsilon_{\mathbf{k}} + e(\nabla_k \epsilon_{\mathbf{k}} \cdot \boldsymbol{\Omega})\mathbf{B}]\,. \tag{2.26}$$

Note that $\epsilon_{\mathbf{k}} = \epsilon^0_{\mathbf{k}} - \boldsymbol{m}_k \cdot \mathbf{B}$. The current density $\boldsymbol{j}$ is given by

$$\boldsymbol{j} = -e\int [d\mathbf{k}] D f(\epsilon_{\mathbf{k}})\dot{\mathbf{r}}\,. \tag{2.27}$$

Focusing on the terms linear in $\mathbf{B}$ we will have

$$\boldsymbol{j} = -e\int [d\mathbf{k}] f(\epsilon_{\mathbf{k}})[\nabla_{\mathbf{k}}\epsilon_{\mathbf{k}} + e(\nabla_k \epsilon_{\mathbf{k}} \cdot \boldsymbol{\Omega})\mathbf{B}] \tag{2.28}$$

$$= -e\int [d\mathbf{k}] f(\epsilon^0)\boldsymbol{v}^0_{\mathbf{k}} + f(\epsilon^0)\nabla_{\mathbf{k}}(-\boldsymbol{m}_{\mathbf{k}} \cdot \mathbf{B}) + \frac{\partial f(\epsilon^0)}{\partial \epsilon^0}(-\boldsymbol{m} \cdot \mathbf{B})\boldsymbol{v}_{\mathbf{k}} + f(\epsilon^0)e(\boldsymbol{v}_{\mathbf{k}} \cdot \boldsymbol{\Omega})\mathbf{B} \tag{2.29}$$

where $\boldsymbol{v}_{\mathbf{k}} = \partial_{\mathbf{k}}\epsilon^0$. The first term gives zero. The second and third term cancel each other after we do integral by parts. Finally we have

$$\boldsymbol{j} = -e^2\int [d\mathbf{k}] f(\epsilon^0_k)(\boldsymbol{v}_{\mathbf{k}} \cdot \boldsymbol{\Omega})\mathbf{B} \tag{2.30}$$

which is zero [76] for the ground state $f(\epsilon^0_{\mathbf{k}})$ or any distribution function depending only on energy. To see that integral over occupied states of $\boldsymbol{\Omega} \cdot \mathbf{v}_{\mathbf{k}}$ vanishes even in the presence of monopole singularities in the Berry curvature, we write, $\mathbf{v}(\mathbf{k}) = \hat{\mathbf{n}} d\varepsilon/dk_\perp$, where $\hat{\mathbf{n}}$ is normal to the surface of constant energy in momentum space and $dk_\perp$ is the separation between two such surfaces whose energy differs by $d\varepsilon$. With this relation, the integral over occupied states can be written,

$$\int [d\mathbf{k}] f(\epsilon^0)\boldsymbol{v}_{\mathbf{k}} \cdot \boldsymbol{\Omega} = \int_{\varepsilon_{min}}^{\mu} d\varepsilon \int_{\varepsilon} dS \boldsymbol{\Omega} \cdot \hat{\mathbf{n}}\,. \tag{2.31}$$

The integral is clearly zero in the absence of singularities in $\boldsymbol{\Omega}$, as in this case $\nabla \cdot \boldsymbol{\Omega} = 0$ for all $\mathbf{k}$. However, the integral still is equal to zero [40] in the presence of singularities such as Weyl nodes, in which case the integral can be written,

$$\int [d\mathbf{k}] f(\epsilon^0)\boldsymbol{v}_{\mathbf{k}} \cdot \boldsymbol{\Omega} = (\mu - \varepsilon_{min}) \sum_n q_n, \tag{2.32}$$

which vanishes as the net monopole charge in the Brillouin zone is zero because of fermion doubling on a lattice [58]. Actually it is required by Bloch's theorem [80] that the " transport coefficient " for the response to a static magnetic field should vanish identically: in equilibrium there are no macroscopic currents in bulk system.

Chapter 3

Gyrotropic magnetic effect and the magnetic moment on the Fermi surface

The current density $\boldsymbol{j}^{\mathbf{B}}$ induced in a clean metal by a magnetic field $\mathbf{B}$ is formulated as the low-frequency limit of natural optical activity, or natural gyrotropy. Working with a multiband Pauli Hamiltonian, we obtain from the Kubo formula a simple expression for $\alpha_{ij}^{\rm gme} = j_i^{\mathbf{B}}/B_j$ in terms of the intrinsic magnetic moment (orbital plus spin) of the Bloch electrons on the Fermi surface. An alternate semiclassical derivation provides an intuitive picture of the effect, and takes into account the influence of scattering processes in dirty metals. This "gyrotropic magnetic effect" is fundamentally different from the chiral magnetic effect driven by the chiral anomaly and governed by the Berry curvature on the Fermi surface, and the two effects are compared for a minimal model of a Weyl semimetal. Like the Berry curvature, the intrinsic magnetic moment should be regarded as a basic ingredient in the Fermi-liquid description of transport in broken-symmetry metals.

3.1 Introduction

When a solid is placed in a static magnetic field the nature of the electronic ground state can change, leading to striking transport effects. A prime example is the integer quantum Hall effect in a quasi two-dimensional (2D) metal in a strong perpendicular field [69]. Novel magnetotransport effects have also been predicted to occur in 3D topological (Weyl) metals, such as an anomalous longitudinal magnetoresistance [56, 65], and the chiral magnetic effect (CME), where an electric pulse $\mathbf{E} \parallel \mathbf{B}$ induces a transient current $\boldsymbol{j} \parallel \mathbf{B}$ [66]; both are related to the chiral anomaly that was originally discussed for Weyl fermions in particle physics [1, 10]. In all these phenomena the role of the static $\mathbf{B}$-field is to modify the equilibrium state, but an $\mathbf{E}$-field is still required to put the electrons out of equilibrium and drive the current (since $\mathbf{E} = -\dot{\mathbf{A}}$, the vector potential is time-dependent even for a static $\mathbf{E}$-field).

Recently, the intriguing proposal was made that a pure **B**-field could drive a dissipationless current in certain Weyl semimetals where isolated band touchings (the "Weyl points," or WPs) of opposite chirality are at different energies [88]. The existence of such an effect was later questioned [71], and the initial interpretation as an *equilibrium* current was discounted. (Indeed, that would a violate a "no-go theorem" attributed to Bloch that forbids macroscopic current in a bulk system in equilibrium [80].) Subsequent theoretical work suggests that the proposed effect can still occur in *transport*, as the current response to a **B**-field oscillating at low frequencies [19, 29, 17, 18].

At present the effect is still widely regarded as being related to the chiral anomaly [19] (or, more generally, to the Berry curvature of the Bloch bands [29, 17, 18, 28]), and is broadly characterized as a type of "CME." We show in this Letter that the experimental implications and microscopic origin of this effect are both very different from the CME (as defined in Ref. [66], consistent with the particle-physics literature [41]). Experimentally, the effect is realized as the low-frequency limit of natural gyrotropy [1] in clean metals (see also Ref. [28]), and we will call it the "gyrotropic magnetic effect" (GME). Both **E** and **B** optical fields drive the gyrotropic current, but at frequencies well below the threshold for interband absorption ($\hbar\omega \ll \epsilon_{\rm gap}$) their separate contributions can be identified. This allows to infer from a low-frequency optical-rotation measurement the reactive current induced by the oscillating **B**-field. The GME is predicted to occur not only in certain Weyl semimetals, but in any optically-active metal (a necessary but not sufficient condition is lack of an inversion center, and a sufficient but not necessary condition is structural chirality [44, 23, 55]).

Existing expressions for the natural gyrotropy current at low frequencies involve the Berry curvature of all the occupied states (and velocities of empty bands) [29, 17, 18, 28], at odds with the notion that transport currents are carried by states near the Fermi level $\epsilon_{\rm F}$. Integrals over all occupied states involving the Berry curvature also appear in calculations of a part of the low-frequency optical activity [61, 35, 86], and of the intrinsic anomalous Hall effect (AHE); in the case of the AHE, a Fermi surface (FS) reformulation exists [30]. We find that the GME is not governed by the chiral anomaly or the Berry curvature, but by the intrinsic magnetic moment of the Bloch states on the FS. Our analysis also takes into account the finite relaxation time τ in real materials, which is shown to weaken the effect at the lowest frequencies. The magnitude of the GME in the clean limit $\omega\tau \gg 1$ is estimated for the optically-active semimetal $SrSi_2$ [38].

[1] The term *natural gyrotropy* refers to the time-reversal-even part of the optical response of a medium at linear order in the wavevector of light [44, 2]. The reactive part gives rise to natural optical activity, and the dissipative part to natural circular dichroism. Gyrotropic effects that are time-reversal-odd and zeroth order in the wavevector of light (e.g., Faraday rotation and magnetic circular dichroism [44]) are not considered in this work.

3.2 Chiral magnetic effect versus gyrotropic magnetic effect

Both effects can be discussed by positing a linear relation between $\boldsymbol{j}$ and $\mathbf{B}$,

$$j_i = \alpha_{ij} B_j. \tag{3.1}$$

Suppose we use linear response to evaluate $\boldsymbol{\alpha}$ for a clean metal, describing the $\mathbf{B}$-field in terms of a vector potential that depends on both $\mathbf{q}$ and ω. The result will depend on the order in which the $\mathbf{q} \to 0$ and $\omega \to 0$ limits are taken [19, 29, 17], much as the compressibility and conductivity are different limits of electrical response. The CME tensor $\boldsymbol{\alpha}^{\rm cme}$ can be obtained from Eq. (3.1) in the equilibrium or *static* limit of the magnetic field (setting $\omega = 0$ before sending $\mathbf{q} \to 0$), with an additional step needed to describe the $\mathbf{E}$-field pulse. The GME tensor $\boldsymbol{\alpha}^{\rm gme}$ is extracted directly from Eq. (3.1) in the transport or *uniform* limit (sending $\mathbf{q} \to 0$ before $\omega \to 0$) that describes conductivities in experiment. (Here "$\omega \to 0$" means $\hbar\omega \ll \epsilon_{\rm gap}$, but note that $\omega\tau \gg 1$ because the clean limit $\tau \to \infty$ is assumed; effects caused by finite relaxation times in dirty samples will be discussed later.) Only $\boldsymbol{\alpha}^{\rm gme}$ is a material property, since the details of the $\mathbf{E}$-field pulse producing nonequilibrium are missing from $\boldsymbol{\alpha}^{\rm cme}$. Below we derive microscopic expressions for both.

3.2.1 Chiral magnetic effect

The tensor $\boldsymbol{\alpha}$ calculated in the static limit is isotropic, $\alpha_{ij} = \alpha^{\rm stat}\delta_{ij}$, with

$$\alpha^{\rm stat} = -\frac{e^2}{\hbar} \sum_n \int [d\mathbf{k}]\, f^0_{\mathbf{k}n} \left(\mathbf{v}_{\mathbf{k}n} \cdot \boldsymbol{\Omega}_{\mathbf{k}n}\right) = 0\,, \tag{3.2}$$

where $[d\mathbf{k}] = d^3k/(2\pi)^3$, the integral is over the Brillouin zone, $f^0_{\mathbf{k}n} = f(\epsilon_{\mathbf{k}n})$ is the equilibrium occupation factor, $\mathbf{v}_{\mathbf{k}n} = \boldsymbol{\partial}_{\hbar\mathbf{k}}\epsilon_{\mathbf{k}n}$ is the band velocity, $\boldsymbol{\Omega}_{\mathbf{k}n} = -{\rm Im}\langle \boldsymbol{\partial}_{\mathbf{k}} u_{\mathbf{k}n}| \times |\boldsymbol{\partial}_{\mathbf{k}} u_{\mathbf{k}n}\rangle$ is the Berry curvature, and $-e$ is the electron charge. Equation (3.2) was derived in Ref. [87] using the semiclassical formalism [75], and we obtain the same result from linear response. The fact that $\alpha^{\rm stat}$ vanishes (see below) is in accord with Bloch's theorem [80].

To turn the above "quasiresponse" into $\alpha^{\rm cme}$, let us recast Eq. (3.2) as a FS integral. Integrating by parts produces two terms. The one containing $\boldsymbol{\partial}_{\mathbf{k}} \cdot \boldsymbol{\Omega}_{\mathbf{k}n}$ picks up monopole contributions from the occupied WPs, and vanishes because each WP appears twice with opposite signs [27]. In the remaining term we write $\boldsymbol{\partial}_{\mathbf{k}} f^0 = -\hat{\mathbf{v}}_{\rm F}\delta^3(\mathbf{k} - \mathbf{k}_{\rm F})$ with $\hat{\mathbf{v}}_{\rm F}$ the FS normal at $\mathbf{k}_{\rm F}$, and introduce the Chern number $C_{na} = (1/2\pi)\int_{S_{na}} dS\,(\hat{\mathbf{v}}_{\rm F} \cdot \boldsymbol{\Omega}_{\mathbf{k}n})$ of the a-th Fermi sheet S_{na} in band n [30, 27]. After assigning different chemical potentials to different sheets to account for the effect of the $\mathbf{E}$-field pulse Eq. (3.2) becomes $\alpha^{\rm cme} = -(e^2/h^2)\sum_{n,a} \mu_{na} C_{na}$, leading to the current density $\boldsymbol{j} = \alpha^{\rm cme}\mathbf{B}$ [66, 80]. In equilibrium $\mu_{na} = \epsilon_{\rm F}$, and using $\sum_{n,a} C_{na} = 0$ we find $\boldsymbol{j} = 0$, as per Eq. (3.2).

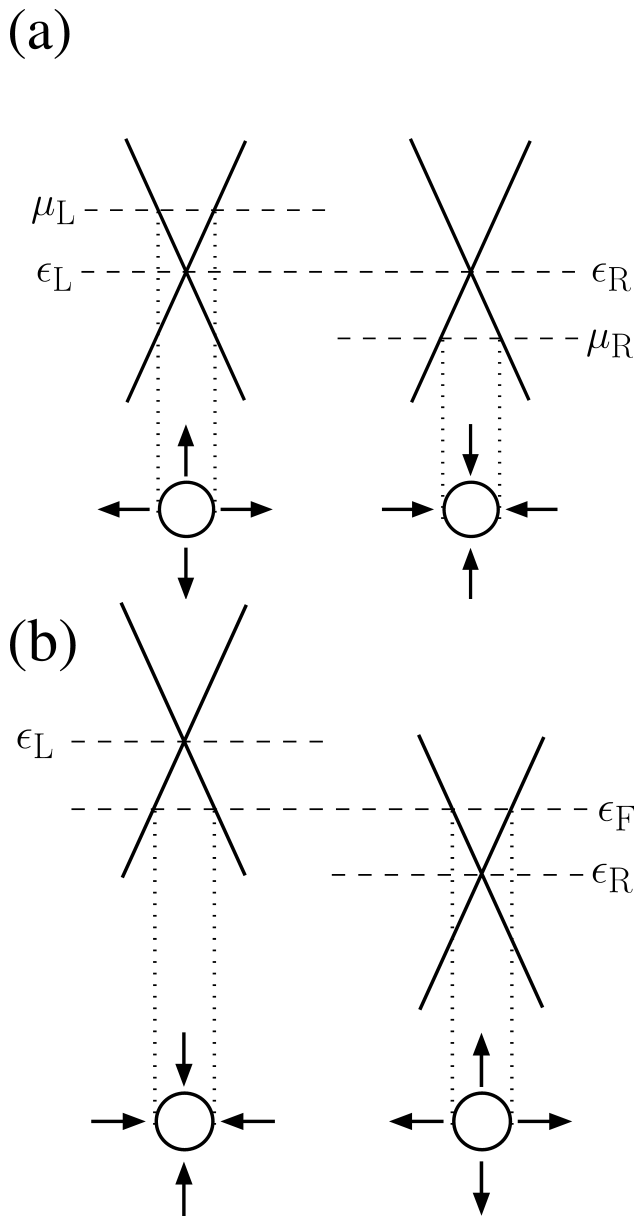

Figure 3.1: (a) Chiral magnetic effect in a T-broken Weyl semimetal in a static $\mathbf{B}$-field. The left- and right-handed Weyl nodes are at the same energy $\epsilon_{\rm L} = \epsilon_{\rm R}$, but the enclosing Fermi pockets are not in chemical equilibrium ($\mu_{\rm L} \neq \mu_{\rm R}$) due to the application of an $\mathbf{E} \parallel \mathbf{B}$ pulse, and this drives the current [Eq. (3.3)]. (b) Gyrotropic magnetic effect. P symmetry is now broken along with T, leading to $\epsilon_{\rm L} \neq \epsilon_{\rm R}$. The Fermi pockets are in chemical equilibrium, $\mu_{\rm L} = \mu_{\rm R} = \epsilon_{\rm F}$, and an oscillating $\mathbf{B}$-field drives the current [Eq. (3.17)]. The bottom of each panel shows the Fermi pockets, and the arrows represent the Fermi velocities.

For a Weyl semimetal with two Fermi pockets with $C = +1$ and $C = -1$ placed at slightly different chemical potentials $\mu_{\rm L}$ and $\mu_{\rm R}$ [2] [Fig. 3.1(a)], a current develops,

$$\boldsymbol{j} = (e^2/h^2)\mathbf{B}(\mu_{\rm R} - \mu_{\rm L})\,. \tag{3.3}$$

3.2.2 Gyrotropic magnetic effect

Symmetry considerations already suggest a link between the GME and natural gyrotropy. Both $\boldsymbol{j}$ and $\mathbf{B}$ are odd under time reversal T, and $\boldsymbol{j}$ is odd under spatial inversion P while $\mathbf{B}$ is P-even, and so according to Eq. (3.1) the GME is T-even and P-odd, same as natural gyrotropy [**Note1**].

To make the connection precise, consider the current density induced by a monochromatic optical field $\mathbf{A}(t,\mathbf{r}) = \mathbf{A}(\omega,\mathbf{q})e^{i(\mathbf{q}\cdot\mathbf{r}-\omega t)}$ at first order in $\mathbf{q}$,

$$j_i(\omega,\mathbf{q}) = \Pi_{ijl}(\omega)A_j(\omega,\mathbf{q})q_l\,. \tag{3.4}$$

The T-even part $\Pi^{\rm A}_{ijl}$ of the response tensor is antisymmetric under $i \leftrightarrow j$. It has nine independent components, and can be repackaged as a rank-2 tensor using [34, 48]

$$\Pi^{\rm A}_{ijl} = i\varepsilon_{ilp}\alpha^{\rm gme}_{jp} - i\varepsilon_{jlp}\alpha^{\rm gme}_{ip} \tag{3.5a}$$

$$\alpha^{\rm gme}_{ij} = \frac{1}{4i}\varepsilon_{jlp}\left(\Pi^{\rm A}_{lpi} - 2\Pi^{\rm A}_{ilp}\right)\,. \tag{3.5b}$$

At nonabsorbing frequencies $\boldsymbol{\alpha}^{\rm gme}(\omega)$ is real and $\boldsymbol{\Pi}^{\rm A}(\omega)$ is purely imaginary, but otherwise both are complex.

From now on we assume $\hbar\omega \ll \epsilon_{\rm gap}$, so that only intraband absorption can occur. In this regime $\boldsymbol{\alpha}^{\rm gme}$ satisfies

$$j^{\mathbf{B}}_i = -i\omega P^{\mathbf{B}}_i = \alpha^{\rm gme}_{ij}B_j \tag{3.6a}$$

$$M^{\mathbf{E}}_i = -(i/\omega)\alpha^{\rm gme}_{ji}E_j\,, \tag{3.6b}$$

where $\mathbf{E} = i\omega\mathbf{A}$ and $\mathbf{B} = i\mathbf{q}\times\mathbf{A}$, and $\mathbf{P}^{\mathbf{B}}$ and $\mathbf{M}^{\mathbf{E}}$ are oscillating moments induced by $\mathbf{B}$ and $\mathbf{E}$ respectively. The gyrotropic current $j^{\rm g}_i = \Pi^{\rm A}_{ijl}A_jq_l = j^{\mathbf{B}}_i + j^{\mathbf{E}}_i$ has contributions from both $\mathbf{B}$ and $\mathbf{E}$, with $\boldsymbol{j}^{\mathbf{B}}$ given by Eq. (3.6a) and $\boldsymbol{j}^{\mathbf{E}} = i\mathbf{q}\times\mathbf{M}^{\mathbf{E}}$. The dissipated power is $\mathrm{Re}(\boldsymbol{j}^{\rm g}\cdot\mathbf{E}^*)/2 = \omega\epsilon_{jlp}q_l\mathrm{Im}(\alpha^{\rm gme}_{ij})\mathrm{Re}(A^*_iA_p)$, confirming that $\mathrm{Re}\,\boldsymbol{\alpha}^{\rm gme}$ and $\mathrm{Im}\,\boldsymbol{\alpha}^{\rm gme}$ control the reactive and dissipative gyrotropic responses respectively.

In the long-wavelength limit Eq. (3.6a) describes a transport current induced by $\mathbf{B}$ in an optically-active metal (the *direct* GME), and Eq. (3.6b) describes a macroscopic magnetization induced by $\mathbf{E}$; the dc limit of this *inverse* GME has been previously discussed for metals with helical crystal structures [83].

[2] With our sign convention for the Berry curvature, a right-handed WP acts as a source in the lower band and as a sink in the upper band [75]. An enclosing pocket, either electron-like or hole-like, has Chern number $C = -1$.

To derive Eq. (3.6), consider a finite sample of size L. Using Eq. (20) of Ref. [48] for $\sigma^{\rm A}_{ijl} = (1/i\omega)\Pi^{\rm A}_{ijl}$ we find [3]

$$\alpha^{\rm gme}_{ij} = (\omega/2i)\left(\chi^{\rm em}_{ij} - \chi^{\rm me}_{ji}\right) + (\text{E. Q. terms})\,. \tag{3.7}$$

"E. Q." denotes electric quadrupole terms that keep $\boldsymbol{\alpha}^{\rm gme}$ origin-independent at higher frequencies [16, 48], but do not contribute to $\boldsymbol{j}^{\mathbf{B}}$ or $\mathbf{M}^{\mathbf{E}}$ when $\hbar\omega \ll \epsilon_{\rm gap}$, as they are higher-order in ω than the first term. The low-frequency gyrotropic response is controlled by the magnetoelectric susceptibilities $\chi^{\rm em}_{ij} = \partial P_i/\partial B_j$ and $\chi^{\rm me}_{ij} = \partial M_i/\partial E_j$. The dynamic polarization $P^{\mathbf{B}}_i$ can be decomposed into T-even and T-odd parts $(1/2)(\chi^{\rm em}_{ij} - \chi^{\rm me}_{ji})B_j$ and $(1/2)(\chi^{\rm em}_{ij} + \chi^{\rm me}_{ji})B_j$[4], and Eq. (3.6a) corresponds to the former. Similarly, Eq. (3.6b) gives the T-even part of the magnetization induced by $\mathbf{E}$. (The T-odd part of the magnetoelectric susceptibilities describes the linear magnetoelectric effect in insulators such as Cr_2O_3.)

In brief, the GME is the low-frequency limit of natural gyrotropy in P-broken metals, in much the same way that the AHE is the transport limit of Faraday rotation in T-broken metals. While the intrinsic AHE is governed by the geometric Berry curvature [75, 30] and becomes quantized by topology in Chern insulators, the GME is controlled by a nongeometric quantity, the intrinsic magnetic moment of the Bloch states [5].

To establish this result let us return to periodic crystals and derive a bulk formula for $\boldsymbol{\alpha}^{\rm gme}$ at $\hbar\omega \ll \epsilon_{\rm gap}$. From Kubo linear response in the uniform limit we obtain [**Note2**]

$$\begin{aligned}\Pi^{\rm A}_{ijl} &= \frac{e^2\omega\tau}{1-i\omega\tau}\sum_n\int[d\mathbf{k}]\,\frac{\partial f}{\partial\epsilon_{\mathbf{k}n}}\Big[-\frac{g_s}{2m_e}\varepsilon_{ipl}v_{\mathbf{k}n,j}S_{\mathbf{k}n,p}\\ &+ \frac{v_{\mathbf{k}n,i}}{\hbar}\mathrm{Im}\langle\partial_j u_{\mathbf{k}n}|H_{\mathbf{k}} - \epsilon_{\mathbf{k}n}|\partial_l u_{\mathbf{k}n}\rangle - (i\leftrightarrow j)\Big].\end{aligned} \tag{3.8}$$

(The calculation was carried out for a clean metal where formally $\tau = 1/\eta$ and $\eta \to 0^+$ [4]. Alternately one could retain a finite τ to give a phenomenological relaxation time in dirty metals, and indeed the semiclassical relaxation-time calculation to be presented shortly gives the same Drude-like dependence on $\omega\tau$ as Eq. (3.8).) $\mathbf{S}_{\mathbf{k}n}$ is the expectation value of the spin $\mathbf{S} = (\hbar/2)\boldsymbol{\sigma}$ of a Bloch state, $g_s \simeq 2$ is the spin g-factor of the electron, and m_e is the electron mass. Inserting Eq. (3.8) in Eq. (3.5b) gives

$$\alpha^{\rm gme}_{ij} = \frac{i\omega\tau e}{1-i\omega\tau}\sum_n\int[d\mathbf{k}]\,(\partial f/\partial\epsilon_{\mathbf{k}n})v_{\mathbf{k}n,i}m_{\mathbf{k}n,j}\,, \tag{3.9}$$

where $\mathbf{m}_{\mathbf{k}n} = -(eg_s/2m_e)\mathbf{S}_{\mathbf{k}n} + \mathbf{m}^{\rm orb}_{\mathbf{k}n}$ is the magnetic moment of a Bloch electron, whose orbital part is [75]

$$\mathbf{m}^{\rm orb}_{\mathbf{k}n} = \frac{e}{2\hbar}\mathrm{Im}\langle\boldsymbol{\partial}_{\mathbf{k}}u_{\mathbf{k}n}|\times(H_{\mathbf{k}} - \epsilon_{\mathbf{k}n})|\boldsymbol{\partial}_{\mathbf{k}}u_{\mathbf{k}n}\rangle\,. \tag{3.10}$$

[3] To recover the bulk result from Eq. (3.7), the $L\to\infty$ limit should be taken faster than the $\omega\to 0$ limit, consistent with the order of limits discussed earlier for transport.

[4] This decomposition is obtained by invoking the Onsager relation $\chi^{\rm em}_{ij}(\omega)\big|_{-\mathbf{B}_{\rm ext}} = -\,\chi^{\rm me}_{ji}(\omega)\big|_{\mathbf{B}_{\rm ext}}$ [51].

[5] Here the term *geometric* refers to the intrinsic geometry of the Bloch-state fiber bundle. The orbital moment of Bloch electrons can be considered geometric in a different sense: it is the imaginary part of a complex tensor whose real part gives the inverse effective mass tensor, i.e., the curvature of band dispersions [26].

At zero temperature, we can replace $\partial f/\partial\epsilon_{\mathbf{k}n}$ in Eq. (3.9) with $-\delta^3(\mathbf{k}-\mathbf{k}_\mathrm{F})/\hbar|\mathbf{v}_{\mathbf{k}n}|$ to obtain the FS formula

$$\alpha_{ij}^{\mathrm{gme}} = \frac{e}{(2\pi)^2 h}\frac{i\omega\tau}{i\omega\tau-1}\sum_{n,a}\int_{S_{na}} dS\,\hat{v}_{\mathrm{F},i} m_{\mathbf{k}n,j}\,. \tag{3.11}$$

A nonzero $\mathbf{m}_{\mathbf{k}n}$ requires broken PT symmetry, but the GME can only occur if P is broken: with P symmetry present $\mathbf{m}_{-\mathbf{k},n} = \mathbf{m}_{\mathbf{k}n}$ and $\hat{\mathbf{v}}_\mathrm{F}(-\mathbf{k}_\mathrm{F}) = -\hat{\mathbf{v}}_\mathrm{F}(\mathbf{k}_\mathrm{F})$, leading to $\boldsymbol{\alpha}^{\mathrm{gme}} = 0$. Without spin-orbit coupling, only the orbital moment contributes.

Equations (3.6) and (3.11) are our main results. The GME is fully controlled by the bulk FS and vanishes trivially for insulators, contrary to the AHE where the FS formulation misses possible quantized contributions [30].

According to Eq. (3.11), the reactive response $\mathrm{Re}\,\boldsymbol{\alpha}^{\mathrm{gme}}$ is suppressed by scattering when $\omega \ll 1/\tau$. It increases with ω, and levels off for $\omega \gg 1/\tau$ (satisfying this condition without violating $\hbar\omega \ll \epsilon_{\mathrm{gap}}$ requires sufficiently clean samples). The opposite is true for the dissipative response $\mathrm{Im}\,\boldsymbol{\alpha}^{\mathrm{gme}}$, which drops to zero at $\omega \gg 1/\tau$ and becomes strongest at $\omega \ll 1/\tau$. In this lowest-frequency limit $\boldsymbol{j}^{\mathbf{B}} \to 0$, and Eqs. (3.6b) and (3.9) for the induced magnetization reduce to the expression in Ref. [83]. Thus, in the dc limit only a dissipative inverse GME occurs.

3.3 Semiclassical picture of the gyrotropic magnetic effect

Our discussion of the GME assumed from the outset $\hbar\omega \ll \epsilon_{\mathrm{gap}}$. Since this is the regime where the semiclassical description of transport in metals holds [6], it is instructive to rederive Eqs. (3.6) and (3.11) by solving the Boltzmann equation. This provides an intuitive picture of the GME and its modification by scattering processes. The key ingredient beyond previous semiclassical approaches [61, 35, 86] is the correction to the band velocity (as opposed to the Berry-curvature anomalous velocity) in the presence of a magnetic field [75]: $\mathbf{v}_{\mathbf{k}n} = \boldsymbol{\partial}_{\hbar\mathbf{k}}\tilde{\epsilon}_{\mathbf{k}n}$, where $\tilde{\epsilon}_{\mathbf{k}n} = \epsilon_{\mathbf{k}n} - \mathbf{m}_{\mathbf{k}n}\cdot\mathbf{B}$.

In a static $\mathbf{B}$-field, the conduction electrons reach a new equilibrium state with $f^0_{\mathbf{k}n}(\mathbf{B}) = f(\tilde{\epsilon}_{\mathbf{k}n})$ as the distribution function [17], and the current vanishes according to Eq. (3.2). Under oscillating fields $\mathbf{E}$, $\mathbf{B} \propto e^{i(\mathbf{q}\cdot\mathbf{r}-\omega t)}$ the electrons are in an excited state with a distribution function $g_{\mathbf{k}n}(t,\mathbf{r})$ which we find by solving the Boltzmann equation in the relaxation-time approximation,

$$\partial_t g_{\mathbf{k}n} + \dot{\mathbf{r}}\frac{\partial g_{\mathbf{k}n}}{\partial\mathbf{r}} + \dot{\mathbf{k}}\frac{\partial g_{\mathbf{k}n}}{\partial\mathbf{k}} = -\left[g_{\mathbf{k}n} - f^0_{\mathbf{k}n}(\mathbf{B})\right]/\tau\,, \tag{3.12}$$

where τ is the relaxation time to return to the instantaneous equilibrium state described by $f^0_{\mathbf{k}n}(\mathbf{B}(t,\mathbf{r}))$ (for a slow spatial variation of $\mathbf{B}$). Using the semiclassical equations [75], the distribution function to linear order in $\mathbf{E}$ and $\mathbf{B}$ is $g_{\mathbf{k}n}(t,\mathbf{r}) = f^0_{\mathbf{k}n}(\mathbf{B}(t,\mathbf{r})) + f^1_{\mathbf{k}n}(t,\mathbf{r})$ with

$$f^1_{\mathbf{k}n} = \frac{\partial f/\partial\epsilon_{\mathbf{k}n}}{1-\frac{\mathbf{q}}{\omega}\cdot\mathbf{v}_{\mathbf{k}n}+\frac{i}{\omega\tau}}\left[\mathbf{m}_{\mathbf{k}n}\cdot\mathbf{B} + (ie/\omega)\mathbf{E}\cdot\mathbf{v}_{\mathbf{k}n}\right], \tag{3.13}$$

which at $\omega\tau \gg 1$ reduces to the result in Ref. [17].

As the current associated with $f^0_{\mathbf{k}n}(\mathbf{B})$ vanishes, the current induced by an oscillating $\mathbf{B}$-field is obtained by multiplying the first term in Eq. (3.13) with the band velocity. The result in the long-wavelength limit is

$$\boldsymbol{j}^{\mathbf{B}} = \frac{ie\omega\tau}{1-i\omega\tau}\sum_n \int [d\mathbf{k}]\,(\partial f/\partial\epsilon_{\mathbf{k}n})\,\mathbf{v}_{\mathbf{k}n}\,(\mathbf{m}_{\mathbf{k}n}\cdot\mathbf{B})\,, \tag{3.14}$$

in agreement with Eqs. (3.6a) and (3.9). Conversely, inserting the second term of Eq. (3.13) in the bulk expression for $\mathbf{M} = \mathbf{M}^{\rm spin} + \mathbf{M}^{\rm orb}$ [75] leads to Eqs. (3.6b) and (3.9) for the magnetization induced by an oscillating $\mathbf{E}$-field.

3.4 Applications in topological semimetals

Consider a situation where only two bands are close to $\epsilon_{\rm F}$, and couplings to more distant bands can be neglected when evaluating the orbital moment on the FS (for simplicity, we focus here on the orbital contribution to the GME in the clean limit). The 2×2 Hamiltonian written in the basis of the identity matrix and the three Pauli matrices is $H_{\mathbf{k}} = \bar{\epsilon}_{\mathbf{k}}\mathbb{1} + \mathbf{d}_{\mathbf{k}}\cdot\boldsymbol{\sigma}$, with eigenvalues $\epsilon_{\mathbf{k}t} = \bar{\epsilon}_{\mathbf{k}} + t d_{\mathbf{k}}$ where $t = \pm 1$ and $d_{\mathbf{k}} = |\mathbf{d}_{\mathbf{k}}|$. Equation (3.99) becomes

$$m^{\rm orb}_{\mathbf{k}t,i} = -\frac{e}{\hbar}\varepsilon_{ijl}\frac{1}{2d^2_{\mathbf{k}}}\mathbf{d}_{\mathbf{k}}\cdot(\partial_j\mathbf{d}_{\mathbf{k}}\times\partial_l\mathbf{d}_{\mathbf{k}})\,. \tag{3.15}$$

For orientation we study a minimal model for a Weyl semimetal where the FS consists of two pockets surrounding isotropic WPs of opposite chirality. We allow the WPs to be at different energies (this requires breaking P in addition to T), but $\epsilon_{\rm F}$ is assumed close to both [Fig. 3.1(b)]. Near each WP the Hamiltonian is $H_{\mathbf{k}\nu} = \epsilon_\nu\mathbb{1} + \chi_\nu\hbar v_{\rm F}\mathbf{k}\cdot\boldsymbol{\sigma}$, where ν labels the WP, ϵ_ν and $\chi_\nu = \pm 1$ are its energy and chirality (positive means right-handed), $\mathbf{k}$ is measured from the WP, and $v_{\rm F}$ is the Fermi velocity. From Eq. (3.15) $\mathbf{m}^{\rm orb}_{\mathbf{k}\nu} = -\chi_\nu(ev_{\rm F}/2k)\hat{\mathbf{k}}$, and only the trace piece $\overline{\alpha}^{\rm gme} = \left(\sum_i \alpha^{\rm gme}_{ii}\right)/3$ survives is Eq. (3.11); in the clean limit each pocket contributes

$$\overline{\alpha}^{\rm gme}_\nu = \mp\frac{1}{3}\frac{e^2}{h^2}\chi_\nu\hbar v_{\rm F}k_{\rm F} = \frac{1}{3}\frac{e^2}{h^2}\chi_\nu(\epsilon_\nu - \epsilon_{\rm F})\,, \tag{3.16}$$

where the minus (plus) sign in the middle expression corresponds to $\epsilon_\nu < \epsilon_{\rm F}$ ($\epsilon_\nu > \epsilon_{\rm F}$). Summing over ν and using $\sum_\nu \chi_\nu = 0$ [57] gives $\overline{\alpha}^{\rm gme} = (e^2/3h^2)\sum_\nu \chi_\nu\epsilon_\nu$. For a minimal model $\nu = {\rm R}, {\rm L}$, and the GME current is

$$\boldsymbol{j}^{\mathbf{B}} = (e^2/3h^2)(\epsilon_{\rm R} - \epsilon_{\rm L})\mathbf{B}\,. \tag{3.17}$$

Equation (3.17) looks deceptively similar to Eq. (3.3) for the CME. The prefactor is different, but the key difference is in the meaning of the various quantities, and in their respective roles. To stress this point, we have in both equations placed the "force" that drives

the current at the end, after the equilibrium parameter that enables the effect. The GME current is driven by the oscillating **B**-field, while $\epsilon_{\rm L}$ and $\epsilon_{\rm R}$ are bandstructure parameters, with $\epsilon_{\rm R} - \epsilon_{\rm L}$ reflecting the degree of structural symmetry breaking that allows the effect to occur. Equation (3.3) is "universal" because of the topological nature of the FS integral involved, while Eq. (3.17) is for spherical pockets surrounding isotropic Weyl nodes. For generic two-band models [6], the non-FS expression found in Refs. [29, 17] for the orbital contribution to $\overline{\alpha}^{\rm gme}$ can be recovered from Eq. (3.9) in the clean limit.

We emphasize that breaking T is not required for the GME. If T is present (and P broken), the minimum number of WPs is four, not two [84]. In the class of T-symmetric Weyl materials so far discovered, T relates WPs of the same chirality and energy. Mirror symmetries connect WPs of opposite chirality so that $\boldsymbol{j}^{\mathbf{B}} = 0$, as expected since these symmetries tend to exclude optical activity [23, 55]. Fortuitously, the predicted Weyl material $SrSi_2$ has misaligned WPs of opposite chirality due to broken mirror symmetry [38]. Its optical activity coefficient ρ can be estimated from the energy splitting between WPs. Neglecting spin contributions that were not included in Eq. (3.17), each WP pair contributes.

$$\rho = (2\alpha/3hc)\,(\epsilon_{\rm L} - \epsilon_{\rm R})\ , \tag{3.18}$$

with α the fine-structure constant and c the speed of light. The calculated splitting $|\epsilon_{\rm L} - \epsilon_{\rm R}| \sim 0.1$ eV [38] gives $|\rho| \sim 0.4$ rad/mm per node pair, about the same as $|\rho| = 0.328$ rad/mm for quartz at $\lambda = 0.63$ μm [55]. This should be measurable in a frequency range from the infrared (above which the semiclassical assumptions break down) down to $1/\tau$, which depends on crystal quality. When $\epsilon_{\rm L} = \epsilon_{\rm R}$ the natural optical activity vanishes in equilibrium, but a nonequilibrium gyrotropic effect can still occur due to the chiral anomaly [35]. In polar metals, the tensor $\boldsymbol{\alpha}^{\rm gme}$ acquires am antisymmetric part (equivalent to a polar vector $\boldsymbol{\delta}$) that does not contribute to optical rotation, but which leads to a *transverse* GME of the form $\mathbf{M}^{\mathbf{E}} \propto \mathbf{E} \times \boldsymbol{\delta}$.

[6] In anisotropic models the traceless part of $\boldsymbol{\alpha}^{\rm gme}$ is generally nonzero. For any number of bands, it includes the traceless piece found previously [86], and the full tensor satisfies the microscopic constraint from time-reversal invariance previously shown for one piece [86].

3.5 Supplemental material for gyrotropic magnetic effect

3.5.1 Kubo formula for natural gyrotropy in crystals

The electronic structure of the crystal is treated at the independent-particle level taking into account the spin-orbit interaction. The Pauli Hamiltonian has the form [13, 79]

$$\mathcal{H}_0 = \frac{p^2}{2m_e} + V(\mathbf{r}) + \frac{\hbar}{4m_e^2}\left(\boldsymbol{\partial}_{\mathbf{r}} V \times \mathbf{p}\right)\cdot\boldsymbol{\sigma}\,, \tag{3.19}$$

where $V(\mathbf{r}) = V(\mathbf{r}+\mathbf{R})$ is the periodic crystalline potential, $\mathbf{p}$ is the canonical momentum, and $\boldsymbol{\sigma}$ is the vector of Pauli matrices. The kinematic momentum associated with $\mathcal{H}_0$ is

$$\boldsymbol{\pi} = \frac{m_e}{i\hbar}[\mathbf{r}, \mathcal{H}_0] = \mathbf{p} + \frac{\hbar}{4m_e}\boldsymbol{\sigma}\times\boldsymbol{\partial}_{\mathbf{r}} V\,, \tag{3.20}$$

and it satisfies $[r_i, \pi_j] = [r_i, p_j] = i\hbar\delta_{ij}$.

In the presence of an electromagnetic field with vector potential $\mathbf{A}(\mathbf{r}, t)$ the Hamiltonian becomes [79]

$$\mathcal{H} = \frac{1}{2m_e}(\mathbf{p}+e\mathbf{A})^2 + V(\mathbf{r}) + \frac{\hbar}{4m_e^2}\left[\boldsymbol{\partial}_{\mathbf{r}} V\times(\mathbf{p}+e\mathbf{A})\right]\cdot\boldsymbol{\sigma} + g_S\frac{\mu_{\mathrm{B}}}{2}(\boldsymbol{\partial}_{\mathbf{r}}\times\mathbf{A})\cdot\boldsymbol{\sigma}\,, \tag{3.21}$$

where m_e and $-e$ are the electron mass and charge, $g_s = 2.0023$ is the spin g-factor of the electron, and $\mu_{\mathrm{B}} = e\hbar/2m_e$ is the Bohr magneton. Expanding Eq. (3.21) and comparing with Eq. (3.20) we find $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\mathrm{I}} + \mathcal{O}(A^2)$, where

$$\mathcal{H}_{\mathrm{I}} = \frac{e}{2}\left(\mathbf{v}\cdot\mathbf{A} + \mathbf{A}\cdot\mathbf{v}\right) + \frac{g_s e}{2m_e}(\boldsymbol{\partial}_{\mathbf{r}}\times\mathbf{A})\cdot\mathbf{S}\,. \tag{3.22}$$

Here $\mathbf{v} = \boldsymbol{\pi}/m_e$ is the velocity operator without the field,[79] and $\mathbf{S} = (\hbar/2)\boldsymbol{\sigma}$ is the spin operator. The first and second terms describe the orbital and spin (Zeeman) couplings respectively. The interaction Hamiltonian of Eq. (3.22) neglects orbital terms quadratic in $\mathbf{A}$, which do not contribute to the linear response we are interested in.

Consider an optical field $\mathbf{A}(t,\mathbf{r}) = \mathbf{A}(\omega,\mathbf{q})e^{i\mathbf{q}\cdot\mathbf{r}-i\omega t}$. The current-density operator in the Fourier representation is

$$\boldsymbol{j} = -\frac{e}{2V}\left\{[\mathbf{v}+(e/m_e)\mathbf{A}]e^{-i\mathbf{q}\cdot\mathbf{r}} + e^{-i\mathbf{q}\cdot\mathbf{r}}[\mathbf{v}+(e/m_e)\mathbf{A}]\right\} - i\frac{g_s e}{2m_e V}\mathbf{q}\times\mathbf{S}e^{-i\mathbf{q}\cdot\mathbf{r}}\,, \tag{3.23}$$

where V is the volume of the crystal. The first term is the orbital current, comprising paramagnetic ($\mathbf{v}$) and diamagnetic ($\mathbf{A}$) contributions; [74, 21] the diamagnetic term appears because the total orbital current is given by the velocity operator in the presence of the optical field, $\mathbf{v}^{\mathrm{tot}} = (1/i\hbar)[\mathbf{r}, \mathcal{H}] = \mathbf{v} + (e/m_e)\mathbf{A}$. The last term in Eq. (3.23) is the current density associated with the induced spin magnetization.

Expressing the total current density induced at linear order by the optical field as

$$j_i(\omega,\mathbf{q}) = \Pi_{ij}(\omega,\mathbf{q})\mathbf{A}(\omega,\mathbf{q}) \tag{3.24}$$

we find, following the standard perturbative calculation [21, 33] and setting $\hbar = 1$,

$$\Pi_{ij}(\omega,\mathbf{q}) = -\frac{e^2}{m_e}\sum_n \int [d\mathbf{k}]\, f_{\mathbf{k}n}\delta_{ij} - e^2\sum_{n,m}\int [d\mathbf{k}]\, \frac{f^0_{\mathbf{k}-\mathbf{q}/2,n} - f^0_{\mathbf{k}+\mathbf{q}/2,m}}{\epsilon_{\mathbf{k}-\mathbf{q}/2,n} - \epsilon_{\mathbf{k}+\mathbf{q}/2,m} + \omega + i\eta}\, M_{\mathbf{k}nm,ij}(\mathbf{q})\,, \tag{3.25}$$

where $[d\mathbf{k}] \equiv d^3k/(2\pi)^3$, $f^0_{\mathbf{k}n} = f(\epsilon_{\mathbf{k}n})$ is the occupation of the Bloch eigenstate $|\psi_{\mathbf{k}n}\rangle = e^{i\mathbf{k}\cdot\mathbf{r}}|u_{\mathbf{k}n}\rangle$ in equilibrium, and η is a positive infinitesimal. The first term is the diamagnetic response, and the matrix in the second term is given by

$$M_{\mathbf{k}nm,ij}(\mathbf{q}) = I^*_{\mathbf{k}mn,i}(\mathbf{q})I_{\mathbf{k}mn,j}(\mathbf{q}) \tag{3.26}$$

$$\mathbf{I}_{\mathbf{k}mn}(\mathbf{q}) = \mathbf{I}^{\rm orb}_{\mathbf{k}mn}(\mathbf{q}) + \mathbf{I}^{\rm spin}_{\mathbf{k}mn}(\mathbf{q}) \tag{3.27}$$

$$\mathbf{I}^{\rm orb}_{\mathbf{k}mn}(\mathbf{q}) = \langle\psi_{\mathbf{k}+\mathbf{q}/2,m}|e^{i\mathbf{q}\cdot\mathbf{r}}\mathbf{v} + \mathbf{v}e^{i\mathbf{q}\cdot\mathbf{r}}|\psi_{\mathbf{k}-\mathbf{q}/2,n}\rangle/2 = \langle u_{\mathbf{k}+\mathbf{q}/2,m}|\boldsymbol{\partial}_{\mathbf{k}}H_{\mathbf{k}}|u_{\mathbf{k}-\mathbf{q}/2,n}\rangle \tag{3.28}$$

$$\mathbf{I}^{\rm spin}_{\mathbf{k}mn}(\mathbf{q}) = -(ig_s/2m_e)\langle u_{\mathbf{k}+\mathbf{q}/2,m}|\mathbf{q}\times\mathbf{S}|u_{\mathbf{k}-\mathbf{q}/2,n}\rangle\,. \tag{3.29}$$

In Eq. (3.28) $H_{\mathbf{k}} \equiv e^{-i\mathbf{k}\cdot\mathbf{r}}\mathcal{H}_0 e^{i\mathbf{k}\cdot\mathbf{r}}$, and we used $\boldsymbol{\partial}_{\mathbf{k}}H_{\mathbf{k}} = \mathbf{v}_{\mathbf{k}} \equiv e^{-i\mathbf{k}\cdot\mathbf{r}}\mathbf{v}e^{i\mathbf{k}\cdot\mathbf{r}}$ and $\mathbf{v}_{\mathbf{k}\pm\mathbf{q}/2} = \mathbf{v}_{\mathbf{k}}\pm\mathbf{q}/2m_e$.[13] At $\mathbf{q}=0$, Eq. (3.25) reduces to the Kubo formula for the optical conductivity $\sigma_{ij}(\omega,0) = (1/i\omega)\Pi_{ij}(\omega,0)$ in the electric-dipole approximation.[33, 21] The dissipative (anti-Hermitean) part of Eq. (3.25) corresponds to Eq. (40) in Ref. [48].

At nonabsorbing frequencies the response is purely reactive (i.e., Π_{ij} is Hermitean[51]) and we can set $\eta = 0$ in Eq. (3.25). Optical gyrotropy is described by the antisymmetric part $\Pi^{\rm A}_{ij} = (\Pi_{ij} - \Pi_{ji})/2$ at $\mathcal{O}(q)$.[44] We therefore take

$$\Pi^{\rm A}_{ij}(\omega,\mathbf{q}) = -ie^2\sum_{n,m}\int [d\mathbf{k}]\, \frac{f^0_{\mathbf{k}-\mathbf{q}/2,n} - f^0_{\mathbf{k}+\mathbf{q}/2,m}}{\epsilon_{\mathbf{k}-\mathbf{q}/2,n} - \epsilon_{\mathbf{k}+\mathbf{q}/2,m} + \omega}\, \mathrm{Im}\, M_{\mathbf{k}nm,ij}(\mathbf{q}) \tag{3.30}$$

and Taylor expand it to $\mathcal{O}(q)$ to captural natural gyrotropy,[44]

$$\Pi^{\rm A}_{ij}(\omega,\mathbf{q}) = \Pi^{\rm A}_{ij}(\omega,0) + \Pi^{\rm A}_{ijl}(\omega)q_l + \dots\,. \tag{3.31}$$

Our goal is to calculate $\Pi^{\rm A}_{ijl}(\omega \ll \epsilon_{\rm gap}/\hbar)$, with $\epsilon_{\rm gap}/\hbar$ the threshold for interband absorption. This low-frequency regime where the semiclassical description holds can be viewed the $\omega \to 0$ limit of the Kubo formula (Ref. [6], p. 253). In Sec. 3.5.2 we calculate $\Pi^{\rm A}_{ijl}(\omega\to 0)$ from Eq. (3.30) in the static limit (setting $\omega = 0$ before sending $\mathbf{q}\to 0$) and in the uniform limit (sending $\mathbf{q}\to 0$ before $\omega\to 0$). The uniform-limit results are valid in the clean limit $\omega \gg 1/\tau$ where intraband scattering is negligible; the effects of scattering at lower frequencies are included in Sec. 3.5.3.

For future reference we collect below the expressions for all $\mathbf{q}$-dependent quantities in Eq. (3.30) up to $\mathcal{O}(q)$. In particular, we expand the matrix-element part as

$$\mathrm{Im}\, M_{\mathbf{k}nm,ij}(\mathbf{q}) \simeq \mathrm{Im}\, M_{\mathbf{k}nm,ij}(0) + \mathrm{Im}(M_{\mathbf{k}nm,ijl})q_l \tag{3.32}$$

and consider separately the orbital and spin contributions, and the intraband ($m = n$) and interband ($m \neq n$) parts. Using a simplified notation where $|u_{\mathbf{k}n}\rangle \to |n\rangle$ and $\partial_{k_i} \to \partial_i$, and defining $v_{ni} = \partial_i \epsilon_n$ and $S_{ni} = \langle n|S_i|n\rangle$, we find

$$f^0_{\mathbf{k}-\mathbf{q}/2,n} - f^0_{\mathbf{k}+\mathbf{q}/2,m} \simeq (f^0_n - f^0_m) - (1/2)\left(v_{nl}\partial f/\partial\epsilon_n + v_{nm}\partial f/\partial\epsilon_m\right) q_l \tag{3.33}$$

$$\epsilon_{\mathbf{k}-\mathbf{q}/2,n} - \epsilon_{\mathbf{k}+\mathbf{q}/2,m} + \omega \simeq (\epsilon_n - \epsilon_m + \omega) - (1/2)(v_{nl} + v_{ml})q_l \tag{3.34}$$

$$\mathrm{Im}\, M_{\mathbf{k}nn,ij}(0) = \mathrm{Im}(v_{ni}v_{nj}) = 0 \tag{3.35}$$

$$\mathrm{Im}\, M_{\mathbf{k}n\neq m,ij}(0) = -(\epsilon_n - \epsilon_m)^2\, \mathrm{Im}(\langle n|\partial_i m\rangle\langle m|\partial_j n\rangle) \tag{3.36}$$

$$\mathrm{Im}\, M^{\mathrm{orb}}_{\mathbf{k}nn,ijl} = v_{nj}\mathrm{Im}\langle n|\partial_i H|\partial_l n\rangle - (i \leftrightarrow j) \tag{3.37}$$

$$\begin{aligned}\mathrm{Im}\, M^{\mathrm{orb}}_{\mathbf{k}n\neq m,ijl} = (1/2)(\epsilon_n - \epsilon_m)\mathrm{Im}\,[&-\langle\partial_l n|\partial_i H|m\rangle\langle m|\partial_j n\rangle \\ &+\langle n|\partial_i H|\partial_l m\rangle\langle m|\partial_j n\rangle] - (i \leftrightarrow j)\end{aligned} \tag{3.38}$$

$$\mathrm{Im}\, M^{\mathrm{spin}}_{\mathbf{k}nn,ijl} = -(g_s/2m_e)\varepsilon_{ipl}S_{np}v_{nj} - (i \leftrightarrow j) \tag{3.39}$$

$$\mathrm{Im}\, M^{\mathrm{spin}}_{\mathbf{k}n\neq m,ijl} = -(g_s/2m_e)(\epsilon_n - \epsilon_m)\varepsilon_{ipl}\mathrm{Re}(\langle n|S_p|m\rangle\langle m|\partial_j n\rangle) - (i \leftrightarrow j)\,, \tag{3.40}$$

where we have abbreviated $\partial f/\partial\epsilon|_{\epsilon=\epsilon_{\mathbf{k}n}}$ as $\partial f/\partial\epsilon_n$, and $H_{\mathbf{k}}$ as H.

Some useful identities We list here some identities that will be used in subsequent manipulations. Note that Eq. (3.45) has already been used in connection with Eqs. (3.35)-(3.40) above.

$$\langle n|\partial_i m\rangle = -\langle\partial_i n|m\rangle \tag{3.41}$$

$$\sum_m |\partial_i m\rangle\langle m| = -\sum_m |m\rangle\langle\partial_i m| \tag{3.42}$$

$$(\partial_i H)|m\rangle = (\epsilon_m - H)|\partial_i m\rangle + v_{mi}|m\rangle \tag{3.43}$$

$$\sum_m \langle\partial_i n|H|\partial_l m\rangle\langle m|\partial_j n\rangle = -\sum_m \epsilon_m\langle\partial_i n|m\rangle\langle\partial_l m|\partial_j n\rangle \tag{3.44}$$

$$\langle n|\partial_i H|m\rangle = -(\epsilon_n - \epsilon_m)\langle n|\partial_i m\rangle + v_{ni}\delta_{nm} \tag{3.45}$$

$$\langle n|\partial_i H|\partial_l n\rangle = \sum_m (\epsilon_n - \epsilon_m)\langle\partial_i n|m\rangle\langle m|\partial_l n\rangle + v_{ni}\langle n|\partial_l n\rangle \tag{3.46}$$

$$\langle n|\partial_i H|\partial_l m\rangle = \epsilon_n\langle\partial_i n|\partial_l m\rangle - \langle\partial_i n|H|\partial_l m\rangle + v_{ni}\langle n|\partial_l m\rangle\,. \tag{3.47}$$

Equations (3.41)–(3.43) are obtained by differentiating $\langle n|m\rangle = \delta_{nm}$, the completeness relation $\sum_m |m\rangle\langle m| = 1$, and $H|m\rangle = \epsilon_m|m\rangle$ respectively. Equation (3.44) follows from Eq. (3.42), and Eqs. (3.45)-(3.47) from Eq. (3.43).

3.5.2 Low-frequency natural gyrotropic response without dissipation

Orbital part

Static limit Here $\Pi^{\rm A}_{ijl}$ denotes the orbital part of the purely reactive response of Eq. (3.30) at $\mathcal{O}(q)$, calculated by setting $\omega = 0$ before expanding in powers of $\mathbf{q}$. We decompose it into interband and intraband parts,

$$\Pi^{\rm A}_{ijl} = \Pi^{\rm A,inter}_{ijl} + \Pi^{\rm A,intra}_{ijl} . \tag{3.48}$$

The interband part is calculated from Eqs. (3.30), (3.33), (3.34), (3.36) and (3.38),

$$\begin{aligned}\Pi^{\rm A,inter}_{ijl} = -\frac{ie^2}{2}\sum_{n,m\neq n}\int [d\mathbf{k}]\,{\rm Im}\Big\{ & (v_{nl}\partial f/\partial\epsilon_n + v_{nm}\partial f/\partial\epsilon_m)\,\langle n|\partial_i m\rangle\langle m|\partial_j n\rangle(\epsilon_n-\epsilon_m) \\ & -(f^0_n - f^0_m)(v_{nl}+v_{ml})\langle n|\partial_i m\rangle\langle m|\partial_j n\rangle \\ & +(f^0_n - f^0_m)\,[-\langle\partial_l n|\partial_i H|m\rangle\langle m|\partial_j n\rangle \\ & +\langle n|\partial_i H|\partial_l m\rangle\langle m|\partial_j n\rangle - (i\leftrightarrow j)]\Big\}. \end{aligned} \tag{3.49}$$

The first, second, and third lines correspond to differentiating the occupation factors, energies, and matrix elements in Eq. (3.30) respectively, and Eq. (3.45) was used to cancel an energy denominator in the last term. The summation over m can be extended to include $m = n$, and exchanging dummy indices $n \leftrightarrow m$ in terms containing f_m renders them equal to the corresponding f_n terms. Using Eq. (3.41) in some terms we obtain

$$\begin{aligned}\Pi^{\rm A,inter}_{ijl} = -ie^2\sum_{n,m}\int [d\mathbf{k}]\,\Big\{ & -\frac{\partial f}{\partial\epsilon_n}v_{nl}{\rm Im}(\langle\partial_i n|m\rangle\langle m|\partial_j n\rangle)(\epsilon_n-\epsilon_m) \\ & +f^0_n(v_{nl}+v_{ml}){\rm Im}(\langle\partial_i n|m\rangle\langle m|\partial_j n\rangle) \\ & +f^0_n{\rm Im}\,[-\langle\partial_l n|\partial_i H|m\rangle\langle m|\partial_j n\rangle \\ & +\langle n|\partial_i H|\partial_l m\rangle\langle m|\partial_j n\rangle - (i\leftrightarrow j)]\Big\}. \end{aligned} \tag{3.50}$$

At $\omega = 0$ and for $n = m$, the fraction in Eq. (3.30) becomes $\partial f/\partial\epsilon_n$. Using Eq. (3.37) we find for the intraband part

$$\Pi^{\rm A,intra}_{ijl} = -ie^2\sum_n\int [d\mathbf{k}]\,(\partial f/\partial\epsilon_n)\,v_{nj}\,{\rm Im}\langle n|\partial_i H|\partial_l n\rangle - (i\leftrightarrow j)\,, \tag{3.51}$$

which can be rewritten with the help of Eq. (3.46) as

$$\Pi^{\rm A,intra}_{ijl} = -ie^2\sum_n\int [d\mathbf{k}]\,(\partial f/\partial\epsilon_n)\,{\rm Im}\,[-v_{nj}\langle\partial_l n|m\rangle\langle m|\partial_i n\rangle - (i\leftrightarrow j)]\,(\epsilon_n-\epsilon_m)\,. \tag{3.52}$$

Adding Eqs. (3.50) and (3.52) gives $\Pi^{\rm A}_{ijl}$ as a sum of three types of terms, $\Pi^{\rm A}_{ijl} = T_1 + T_2 + T_3$. They are

$$T_1 = -ie^2\sum_{n,m}\int [d\mathbf{k}]\,\frac{\partial f^0_n}{\partial\epsilon_n}\,{\rm Im}\,[-v_{nl}\langle\partial_i n|m\rangle\langle m|\partial_j n\rangle - v_{nj}\langle\partial_l n|m\rangle\langle m|\partial_i n\rangle$$

$$+ v_{ni}\langle\partial_l n|m\rangle\langle m|\partial_j n\rangle]\,(\epsilon_n - \epsilon_m) \tag{3.53}$$

$$T_2 = -ie^2\sum_{n,m}\int [d\mathbf{k}]\, f_n^0\,\mathrm{Im}\left[\langle n|\partial_i H|\partial_l m\rangle\langle m|\partial_j n\rangle - \langle\partial_l n|\partial_i H|m\rangle\langle m|\partial_j n\rangle - (i\leftrightarrow j)\right] \tag{3.54}$$

$$T_3 = -ie^2\sum_{n,m}\int [d\mathbf{k}]\, f_n^0(v_{nl}+v_{ml})\,\mathrm{Im}(\langle\partial_i n|m\rangle\langle m|\partial_j n\rangle)\,. \tag{3.55}$$

Writing $v_{nl}\partial f/\partial\epsilon_n = \partial_l f_n^0$ in T_1 and integrating by parts yields

$$\begin{aligned} T_1 = -ie^2\sum_{n,m}\int [d\mathbf{k}]\, f_n^0\Big\{ \quad & (\epsilon_n-\epsilon_m)\,\mathrm{Im}\left[-\langle\partial_j n|\partial_l m\rangle\langle m|\partial_i n\rangle + \langle\partial_l n|\partial_j m\rangle\langle m|\partial_i n\rangle\right. \\ & \left. + \langle\partial_j n|\partial_i m\rangle\langle m|\partial_l n\rangle - (i\leftrightarrow j)\right] \\ & +(v_{nl}-v_{ml})\,\mathrm{Im}(\langle\partial_i n|m\rangle\langle m|\partial_j n\rangle) \\ & -(v_{nj}-v_{mj})\,\mathrm{Im}(\langle\partial_i n|m\rangle\langle m|\partial_l n\rangle) \\ & +(v_{ni}-v_{mi})\,\mathrm{Im}(\langle\partial_j n|m\rangle\langle m|\partial_l n\rangle)\Big\}\,. \end{aligned} \tag{3.56}$$

In order to facilitate the collection of terms, we have adopted the following conventions. In $\mathrm{Im}(\langle\partial_a n|\partial_b m\rangle\langle m|\partial_c n\rangle)$ we set $c = i, l$ but never $c = j$, and in $\mathrm{Im}(\langle\partial_a n|m\rangle\langle m|\partial_b n\rangle)$ we choose "$a < b$" where "$i < j < l$."

Expanding the term T_2 using Eq. (3.47) followed by Eq. (3.44),

$$\begin{aligned} T_2 = -ie^2\sum_{n,m}\int [d\mathbf{k}]\, f_n^0\big[\quad & - \quad (v_{nj}+v_{mj})\,\mathrm{Im}(\langle\partial_i n|m\rangle\langle m|\partial_l n\rangle) - \epsilon_n\,\mathrm{Im}(\langle\partial_j n|\partial_l m\rangle\langle m|\partial_i n\rangle) \\ & - \quad \epsilon_m\,\mathrm{Im}(\langle\partial_j n|\partial_l m\rangle\langle m|\partial_i n\rangle) - \epsilon_m\,\mathrm{Im}(-\langle\partial_l n|\partial_j m\rangle\langle m|\partial_i n\rangle \\ & - \quad \langle\partial_j n|\partial_i m\rangle\langle m|\partial_l n\rangle) - (i\leftrightarrow j)\big]\,. \end{aligned} \tag{3.57}$$

Consider the identity obtained by multiplying Eq. (3.42) on the left with $\langle\partial_l n|$ and on the right with $|\partial_j n\rangle$. It implies that the second term in Eq. (3.57) (combined with its "$-(i\leftrightarrow j)$" partner) vanishes upon summing over m, and so $-\epsilon_n$ therein can be changed into ϵ_n. Likewise, we can substitute $-\epsilon_m$ in the second line with $\epsilon_n - \epsilon_m$, to find

$$\begin{aligned} T_2 = -ie^2\sum_{n,m}\int [d\mathbf{k}]\, f_n^0\big[& -(v_{nj}+v_{mj})\,\mathrm{Im}(\langle\partial_i n|m\rangle\langle m|\partial_l n\rangle) \\ & + \quad (\epsilon_n-\epsilon_m)\,\mathrm{Im}\left(\langle\partial_j n|\partial_l m\rangle\langle m|\partial_i n\rangle - \langle\partial_l n|\partial_j m\rangle\langle m|\partial_i n\rangle\right. \\ & - \quad \left.\langle\partial_j n|\partial_i m\rangle\langle m|\partial_l n\rangle) - (i\leftrightarrow j)\big]\,. \end{aligned} \tag{3.58}$$

Adding Eqs. (3.55), (3.56), and (3.58) we obtain, upon invoking the completeness relation,

$$\Pi^{\mathrm{A}}_{ijl} = ie^2\sum_n\int [d\mathbf{k}]\, f_n^0\left[v_{nl}\Omega_{n,ij} - (i\leftrightarrow l) - (j\leftrightarrow l)\right]\,, \tag{3.59}$$

where $\Omega_{n,ij} = -2\mathrm{Im}\langle\partial_i n|\partial_j n\rangle = -\Omega_{n,ji}$ is the Berry curvature. The quantity $[\ldots]$ in the previous equation is totally antisymmetric, and so it can be written as $C_n\varepsilon_{ijl}$, where $C_n = (1/6)\varepsilon_{ijl}[\ldots] = \mathbf{\Omega}_n \cdot \mathbf{v}_n$. Thus,

$$\boxed{\Pi^{\mathrm{A}}_{ijl} = (ie^2/\hbar)\varepsilon_{ijl}\sum_n \int [d\mathbf{k}]\, f^0_n(\mathbf{\Omega}_n \cdot \mathbf{v}_n) = 0} \quad \text{(orbital, static limit)} \tag{3.60}$$

where we have restored $\hbar$. Using $j_i = \Pi^{\mathrm{A}}_{ijl}A_j q_l$ and $B_i = -i\varepsilon_{ijl}A_j q_l$ we arrive at Eq. (2). The vanishing of Eq. (3.60) was demonstrated in Ref. [87], and an alternate proof is given in the main text.

Uniform limit We expand Eq. (3.30) in powers of $\mathbf{q}$ keeping ω finite, and send $\omega \to 0$ at the end. This change in the order of limits compared to the static case does not affect the calculation of the interband term $\Pi^{\mathrm{A,inter}}_{ijl}$, but the intraband term now vanishes. To show this, define $F \equiv f^0_{\mathbf{k}-\mathbf{q}/2,n} - f^0_{\mathbf{k}+\mathbf{q}/2,n}$ and $G \equiv \epsilon_{\mathbf{k}-\mathbf{q}/2,n} - \epsilon_{\mathbf{k}+\mathbf{q}/2,n} + \omega$, so that

$$\Pi^{\mathrm{A,intra}}_{ijl} = -ie^2 \lim_{\omega\to 0}\sum_n \int [d\mathbf{k}] \left[(F/G)|_{\mathbf{q}=0}\, \mathrm{Im}M^{\mathrm{orb}}_{\mathbf{k}nn,ijl} + \partial_l\, (F/G)|_{\mathbf{q}=0}\, \mathrm{Im}M^{\mathrm{orb}}_{\mathbf{k}nn,ij}(0) \right], \tag{3.61}$$

where

$$(F/G)|_{\mathbf{q}=0} = (f^0_n - f^0_n)/(\epsilon_n - \epsilon_n + \omega) = 0\,, \tag{3.62a}$$
$$\partial_l\, (F/G)|_{\mathbf{q}=0} = -(1/\omega)v_{nl}\partial f/\partial\epsilon_n\,, \tag{3.62b}$$

and $\mathrm{Im}M^{\mathrm{orb}}_{\mathbf{k}nn,ij}(0) = 0$ according to Eq. (3.35). Thus $\Pi^{\mathrm{A,intra}}_{ijl} = 0$, and so the response in the uniform limit is purely interband. Since the interband term is independent of the order of limits, and the net response vanishes in the static limit, the uniform-limit response equals minus the static-limit intraband term. Using Eq. (3.43) in Eq. (3.51),

$$\Pi^{\mathrm{A}}_{ijl} = (ie^2/\hbar)\sum_n \int [d\mathbf{k}]\, (\partial f/\partial\epsilon_n) v_{ni}\, \mathrm{Im}\langle\partial_j n|H - \epsilon_n|\partial_l n\rangle - (i \leftrightarrow j)\,. \tag{3.63}$$

Spin part

Static limit We again start with the interband part, and collect $\mathcal{O}(q)$ spin contributions to Eq. (3.30). Since $\mathrm{Im}M_{\mathbf{k}n\neq m,ij}(0)$ is purely orbital [Eq. (3.36)] the only contribution comes from Eq. (3.40), yielding

$$\Pi^{\mathrm{A,inter}}_{ijl} = (ig_s e^2/2m_e)\sum_{n,m} \int [d\mathbf{k}]\, (f^0_n - f^0_m)\varepsilon_{ipl}\, \mathrm{Re}(\langle n|S_p|m\rangle\langle m|\partial_j n\rangle) - (i \leftrightarrow j)\,. \tag{3.64}$$

Exchanging $n \leftrightarrow m$ indices in one term and using the completeness relation gives

$$\begin{aligned}\Pi_{ijl}^{\mathrm{A,inter}} &= (ig_s e^2/2m_e)\sum_n \int [d\mathbf{k}]\, f_n^0 \varepsilon_{ipl}\partial_j S_{np} - (i \leftrightarrow j) \\ &= -(ig_s e^2/2m_e)\sum_n \int [d\mathbf{k}]\, (\partial f/\partial\epsilon_n)\varepsilon_{ipl} v_{nj} S_{np} - (i \leftrightarrow j)\,. \end{aligned} \tag{3.65}$$

For the intraband part we use Eq. (3.39), which leads to the expression above with the opposite sign. Thus,

$$\Pi_{ijl}^{\mathrm{A}} = \Pi_{ijl}^{\mathrm{A,inter}} + \Pi_{ijl}^{\mathrm{A,intra}} = 0\,. \tag{3.66}$$

Uniform limit The steps in the derivation are similar to those carried out in Sec. 3.5.2 for the orbital contribution. The intraband part vanishes, and the interband part is the same as in the static limit, Eq. (3.65). Thus,

$$\Pi_{ijl}^{\mathrm{A}} = -(ig_s e^2/2m_e)\sum_n \int [d\mathbf{k}]\, (\partial f/\partial\epsilon_n)\varepsilon_{ipl} v_{nj} S_{np} - (i \leftrightarrow j)\,. \tag{3.67}$$

3.5.3 Low-frequency natural gyrotropic response with dissipation

The low-frequency ($\hbar\omega \ll \epsilon_{\mathrm{gap}}$)natural gyrotropic response was evaluated in Secs. 3.5.2 and 3.5.2 in the clean limit $\omega\tau \gg 1$. It is well known that at $\mathbf{q} = 0$ the Drude formula for the complex optical conductivity (i.e., including both reactive and dissipative parts) can be recovered heuristically from the intraband part of the Kubo formula (3.25) for a pristine metal, by interpreting the positive infinitesimal η as a scattering rate $1/\tau$. [4] Here we apply the same procedure to extend our calculation of natural gyrotropy in metals in the semiclassical limit to arbitrarily low frequencies compared to $1/\tau$. The first step is to restore η in the denominator of Eq. (3.30),

$$\Pi_{ij}^{\mathrm{A}}(\omega,\mathbf{q}) = -ie^2\sum_{n,m}\int [d\mathbf{k}]\, \frac{f^0_{\mathbf{k}-\mathbf{q}/2,n} - f^0_{\mathbf{k}+\mathbf{q}/2,m}}{\epsilon_{\mathbf{k}-\mathbf{q}/2,n} - \epsilon_{\mathbf{k}+\mathbf{q}/2,m} + \hbar\omega + i\hbar\eta}\,\mathrm{Im}\, M_{\mathbf{k}nm,ij}(\mathbf{q})\,, \tag{3.68}$$

and we have also brought back $\hbar$ for clarity. Multiplying and dividing by $\hbar\omega$ and then using the identity

$$\frac{1}{\hbar\omega(\hbar\omega+\Delta)} = \frac{1}{\Delta}\left(\frac{1}{\hbar\omega} - \frac{1}{\hbar\omega+\Delta}\right) \tag{3.69}$$

with $\Delta = \epsilon_{\mathbf{k}-\mathbf{q}/2,n} - \epsilon_{\mathbf{k}+\mathbf{q}/2,m} + i\hbar\eta$ we find

$$\Pi_{ij}^{\mathrm{A}}(\omega,\mathbf{q}) = -\,ie^2\sum_{n,m}\int [d\mathbf{k}]\, \frac{f^0_{\mathbf{k}-\mathbf{q}/2,n} - f^0_{\mathbf{k}+\mathbf{q}/2,m}}{\epsilon_{\mathbf{k}-\mathbf{q}/2,n} - \epsilon_{\mathbf{k}+\mathbf{q}/2,m} + i\hbar\eta}\,\mathrm{Im}\, M_{\mathbf{k}nm,ij}(\mathbf{q})$$

$$+ ie^2\hbar\omega \sum_{n,m} \int [d\mathbf{k}] \, \frac{f^0_{\mathbf{k}-\mathbf{q}/2,n} - f^0_{\mathbf{k}+\mathbf{q}/2,m}}{\epsilon_{\mathbf{k}-\mathbf{q}/2,n} - \epsilon_{\mathbf{k}+\mathbf{q}/2,m} + i\hbar\eta} \, \frac{\mathrm{Im}\, M_{\mathbf{k}nm,ij}(\mathbf{q})}{\hbar(\omega+i\eta) + \epsilon_{\mathbf{k}-\mathbf{q}/2,n} - \epsilon_{\mathbf{k}+\mathbf{q}/2,m}} \,. \tag{3.70}$$

Consider first the dissipative response, given by the real part of this expression. The semiclassical condition $\hbar\omega \ll \epsilon_{\rm gap}$ excludes interband absorption, and so we can set $m = n$. The contribution from the first line is

$$-\pi e^2 \sum_n \int [d\mathbf{k}] \left(f^0_{\mathbf{k}-\mathbf{q}/2,n} - f^0_{\mathbf{k}+\mathbf{q}/2,n} \right) \delta(\epsilon_{\mathbf{k}-\mathbf{q}/2,n} - \epsilon_{\mathbf{k}+\mathbf{q}/2,n}) \mathrm{Im}\, M_{\mathbf{k}nm,ij}(\mathbf{q}) \,, \tag{3.71}$$

and it vanishes identically because whenever the first factor is nonzero the second factor is zero, and vice-versa. Thus, intraband absorption comes entirely from the second line in Eq. (3.70),

$$\mathrm{Re}\, \Pi^{\rm A}_{ij}(\omega \ll \epsilon_{\rm gap}/\hbar, \mathbf{q}) = \mathrm{Re} \left\{ ie^2\hbar\omega \sum_n \int [d\mathbf{k}] \, \frac{\partial f}{\partial \epsilon_n} \frac{\mathrm{Im}\, M_{\mathbf{k}nn,ij}(\mathbf{q})}{\hbar(\omega+i\eta) + \epsilon_{\mathbf{k}-\mathbf{q}/2,n} - \epsilon_{\mathbf{k}+\mathbf{q}/2,n}} \right\} . \tag{3.72}$$

The dissipative part of natural gyrotropy is given by the $\mathcal{O}(q)$ terms in this equation. Since $\mathrm{Im}\, M_{\mathbf{k}nn,ij}(0) = 0$ according to Eq. (3.35), the only contribution comes from Taylor expanding the numerator with $\mathbf{q} = 0$ in the denominator. Identifying η with $1/\tau$ and assuming it is constant across the Fermi surface (FS) we find

$$\mathrm{Re}\, \Pi^{\rm A}_{ijl}(\omega \ll \epsilon_{\rm gap}/\hbar) = e^2\, \mathrm{Re} \left(\frac{\omega\tau}{1 - i\omega\tau} \right) G_{ijl} \,, \tag{3.73}$$

where

$$G_{ijl} = \sum_n \int [d\mathbf{k}] \, (\partial f/\partial \epsilon_n) \, \mathrm{Im}\, M_{\mathbf{k}nn,ijl} = G^{\rm orb}_{ijl} + G^{\rm spin}_{ijl} \tag{3.74a}$$

$$G^{\rm orb}_{ijl} = (1/\hbar) \sum_n \int [d\mathbf{k}] \, (\partial f/\partial \epsilon_n) v_{ni} \, \mathrm{Im} \langle \partial_j n | H - \epsilon_n | \partial_l n \rangle - (i \leftrightarrow j) \tag{3.74b}$$

$$G^{\rm spin}_{ijl} = -(g_s/2m_e) \sum_n \int [d\mathbf{k}] \, (\partial f/\partial \epsilon_n) \varepsilon_{ipl} v_{nj} S_{np} - (i \leftrightarrow j) \,. \tag{3.74c}$$

To obtain the orbital term we used Eqs. (3.37) and (3.43), and for the spin term we used Eq. (3.39).

The reactive part can be recovered from the Kramers-Krönig relation [7] for $\sigma_{ijl} = (1/i\omega)\, \Pi_{ijl}$, the $\mathcal{O}(q)$ part of the effective conductivity defined in Eq. (3.82) below. The result for the full (complex) response at low frequencies is

$$\boxed{\Pi^{\rm A}_{ijl}(\omega \ll \epsilon_{\rm gap}/\hbar) = e^2 \, \frac{\omega\tau}{1 - i\omega\tau} G_{ijl}} \quad \text{(orbital + spin, uniform limit)} \tag{3.75}$$

[7] Like the Drude optical conductivity, natural gyrotropy at $\hbar\omega \ll \epsilon_{\rm gap}$ satisfies the Kramers-Krönig relations.

which can be viewed as a Drude-like formula for natural gyrotropy in the semiclassical regime. At $\omega\tau \ll 1$ the response becomes purely dissipative and at $\omega\tau \gg 1$ it becomes purely reactive, reducing to the result from the rigorous Kubo-formula calculation in the clean limit, Eqs. (3.63) and (3.67). Equation (7) is the combination of Eqs. (3.74) and (3.75).

3.5.4 Orbital GME in two-band models: comparison with the previous literature

In the main text we obtained an expression, Eq. (17), for the orbital GME current $\boldsymbol{j}^{\mathbf{B}} = \overline{\alpha}^{\mathrm{gme}}\mathbf{B}$ at $\omega\tau \gg 1$ in a Weyl semimetal with two isotropic Weyl points (WPs). Here we consider a generic two-band model and show how to recover, starting from the Fermi-surface formula for $\boldsymbol{\alpha}^{\mathrm{gme}}$, the expression given in Refs. [29, 17] for the trace piece $\overline{\alpha}^{\mathrm{gme}} = \mathrm{tr}(\boldsymbol{\alpha}^{\mathrm{gme}})/3$ (but keeping in mind that in anisotropic models the traceless part is generally nonzero; specific traceless contributions will be considered in Sec. 3.5.5.4).

In two-band models the orbital moment is related to the Berry curvature by $\mathbf{m}^{\mathrm{orb}}_{\mathbf{k}t} = (e/\hbar)td_{\mathbf{k}}\boldsymbol{\Omega}_{\mathbf{k}t}$, [82] in the notation of the the main text. Inserting this expression in Eq. (9) and taking the trace gives, at $\omega\tau \gg 1$,

$$\overline{\alpha}^{\mathrm{gme}} = -\frac{e^2}{3\hbar}\sum_{t=\pm}\int [d\mathbf{k}]\,(\partial f/\partial\epsilon_{\mathbf{k}t})td_{\mathbf{k}}\mathbf{v}_{\mathbf{k}t}\cdot\boldsymbol{\Omega}_{\mathbf{k}t}\,. \tag{3.76}$$

Replacing $\mathbf{v}_{\mathbf{k}t}\partial f/\partial\epsilon_{\mathbf{k}t}$ with $\boldsymbol{\partial}_{\hbar\mathbf{k}}f^0_{\mathbf{k}t}$ and integrating by parts yields two terms; the one containing $\boldsymbol{\partial}_{\mathbf{k}}\cdot\boldsymbol{\Omega}_{\mathbf{k}t}$ vanishes identically (see below), leaving

$$\overline{\alpha}^{\mathrm{gme}} = \frac{e^2}{3\hbar}\sum_{t=\pm}\int [d\mathbf{k}]\, f^0_{\mathbf{k}t}\boldsymbol{\Omega}_{\mathbf{k}t}\cdot\boldsymbol{\partial}_{\hbar\mathbf{k}}(td_{\mathbf{k}})\,. \tag{3.77}$$

Writing $td_{\mathbf{k}}$ as $\epsilon_{\mathbf{k}t} - (\epsilon_{\mathbf{k}+} + \epsilon_{\mathbf{k}-})/2$ and using Eq. (2) to eliminate a term we find

$$\overline{\alpha}^{\mathrm{gme}} = -\frac{e^2}{3\hbar}\sum_{t=\pm}\int [d\mathbf{k}]\, f^0_{\mathbf{k}t}\frac{\mathbf{v}_{\mathbf{k}+}+\mathbf{v}_{\mathbf{k}-}}{2}\cdot\boldsymbol{\Omega}_{\mathbf{k}t}\,. \tag{3.78}$$

The expression in Refs. [29, 17] amounts to 3/2 times Eq. (3.78) minus 1/2 times Eq. (3.76), divided by two. To understand the division by two, recall from Eq. (6a) that $\boldsymbol{\alpha}^{\mathrm{gme}}$ describes the current response to the **B**-field component of the optical field. The **E**- and **B**-fields contribute equal amounts to the component of the gyrotropic current along **B** (see Sec. 3.5.5.1 below), bringing our result in accord with Refs. [29, 17]. While strictly correct, the above expression for $\overline{\alpha}^{\mathrm{gme}}$ in two-band models is not physically transparent. It fails to recognize the FS nature of the GME, and has led to the erroneous identification of the Berry curvature as the key quantity controlling the effect.[29, 17, 18]

Between Eqs. (3.76) and (3.77) we dropped a term containing the divergence of the Berry curvature,

$$\frac{e^2}{3\hbar^2}\sum_{t=\pm}\int [d\mathbf{k}]\, f_{\mathbf{k}t}(\epsilon_{\mathbf{k}t} - \bar{\epsilon}_{\mathbf{k}})\,(\boldsymbol{\partial}_{\mathbf{k}}\cdot\boldsymbol{\Omega}_{\mathbf{k}t})\,. \tag{3.79}$$

The Berry curvature $\mathbf{\Omega}_{\mathbf{k}t} = \boldsymbol{\partial} \times \mathbf{A}_{\mathbf{k}t}$ is divergence-free except at the WPs $\epsilon_{\mathbf{k}t} = \bar{\epsilon}_{\mathbf{k}}$, which act as monopole sources and sinks. The above expression is proportional to the sum over all WPs, and over the two bands, of $f_{\mathbf{k}t}$ times $\epsilon_{\mathbf{k}t} - \bar{\epsilon}_{\mathbf{k}}$ times δ-function singularities. Since $f_{\mathbf{k}t} = 0, 1$ and $\epsilon_{\mathbf{k}t} - \bar{\epsilon}_{\mathbf{k}} = 0$ are the same for the two bands at a WP, the summation only acts on the last factor. But $\sum_{t=\pm} \boldsymbol{\partial}_{\mathbf{k}} \cdot \mathbf{\Omega}_{\mathbf{k}t} = 0$, because each WP contributes twice with opposite signs: once as a source term in one band, and another time as sink in the other band (see, e.g., Eq. (10) in Ref. [27]). Thus, each WP gives a vanishing net contribution to Eq. (3.79), which therefore vanishes identically.

3.5.5 Natural optical activity of metals in the semiclassical limit

3.5.5.1 Gyrotropic response in equilibrium

The GME in metals is conveniently formulated in terms of the dual tensors $\Pi^{\mathrm{A}}_{ijl} = -\Pi^{\mathrm{A}}_{jil}$ and $\alpha^{\mathrm{gme}}_{ij}$, related to one another by Eq. (5). Instead, the gyrotropic response of a bulk medium is usually discussed in terms of the antisymmetric part of the relative permittivity tensor $\epsilon^{\mathrm{A}}_{ij}(\omega, \mathbf{q}) = [\epsilon_{ij}(\omega, \mathbf{q}) - \epsilon_{ji}(\omega, \mathbf{q})]/2$ expanded to $\mathcal{O}(q)$,[44]

$$\epsilon^{\mathrm{A}}_{ij}(\omega, \mathbf{q}) = \epsilon^{\mathrm{A}}_{ij}(\omega, 0) + i\gamma^{\mathrm{A}}_{ijl}(\omega) q_l + \dots \,. \tag{3.80}$$

The T-odd tensor $\epsilon^{\mathrm{A}}_{ij}(\omega, 0)$ describes magneto-optical gyrotropic effects such as Faraday rotation and magnetic circular dichroism, and the T-even tensor $\gamma^{\mathrm{A}}_{ijl}(\omega) = -\gamma^{\mathrm{A}}_{jil}(\omega)$ describes natural gyrotropy (natural optical activity and natural circular dichroism). It is also useful to introduce a dimensionless *gyration tensor* $g_{ij}(\omega)$ dual to $\gamma^{\mathrm{A}}_{ijl}(\omega)$ according to[44]

$$\gamma^{\mathrm{A}}_{ijl} = (c/\omega)\varepsilon_{ijm} g_{ml} \tag{3.81a}$$

$$g_{ij} = (\omega/2c)\varepsilon_{ilm}\gamma^{\mathrm{A}}_{lmj}\,, \tag{3.81b}$$

where c is the speed of light. In order to convert between Π^{A}_{ijl} and $\alpha^{\mathrm{gme}}_{ij}$ on one hand and $\gamma^{\mathrm{A}}_{ijl}$ and g_{ij} on the other, consider the effective conductivity tensor $\sigma_{ij}(\omega, \mathbf{q})$ satisfying[51, 48]

$$j_i(\omega, \mathbf{q}) = \sigma_{ij}(\omega, \mathbf{q}) E_j(\omega, \mathbf{q})\,. \tag{3.82}$$

Using $\mathbf{E} = i\omega\mathbf{A}$ and comparing with Eq. (3.24) gives $\Pi_{ij} = i\omega\sigma_{ij}$. Hence the tensors ϵ_{ij}, σ_{ij}, and Π_{ij} are related by[51]

$$\epsilon_{ij}(\omega, \mathbf{q}) = \delta_{ij} + \frac{i}{\omega\epsilon_0}\sigma_{ij}(\omega, \mathbf{q}) = \delta_{ij} + \frac{1}{\omega^2\epsilon_0}\Pi_{ij}(\omega, \mathbf{q})\,. \tag{3.83}$$

Expanding the antisymmetric part to $\mathcal{O}(q)$ and comparing with Eq. (3.80) yields the relation

$$\gamma^{\mathrm{A}}_{ijl} = -(i/\omega^2\epsilon_0)\Pi^{\mathrm{A}}_{ijl} \tag{3.84}$$

between the two rank-3 gyrotropic tensors. The relation between their duals g_{ij} and $\alpha^{\rm gme}_{ij}$ follows from combining Eqs. (3.81b), (3.84), and (5a). The result is (note the transposed indices)

$$g_{ij} = \frac{1}{\omega c \epsilon_0}\left(\widetilde{\alpha}^{\rm gme}_{ji} - 2\overline{\alpha}^{\rm gme}\delta_{ij}\right) , \tag{3.85}$$

where $\overline{\alpha}^{\rm gme}\delta_{ij}$ and $\widetilde{\alpha}^{\rm gme}_{ij}$ are respectively the trace piece and the traceless part of $\alpha^{\rm gme}_{ij}$.

Let us now specialize to a metal with cubic symmetry or higher, and express the optical activity coefficient ρ (rotation angle per unit transmission length) in terms of the GME tensor. In a medium with such high symmetry the tensors $\Pi^{\rm A}_{ijl}$ and $\gamma^{\rm A}_{ijl}$ are totally antisymmetric. From Eq. (5) we get $\Pi^{\rm A}_{ijl} = -2i\overline{\alpha}^{\rm gme}\varepsilon_{ijl}$ and $\alpha^{\rm gme}_{ij} = \overline{\alpha}^{\rm gme}\delta_{ij}$, Eq. (6) gives $\boldsymbol{j}^{\mathbf{B}} = \boldsymbol{j}^{\mathbf{E}} = \overline{\alpha}^{\rm gme}\mathbf{B}$, and finally $\gamma^{\rm A}_{ijl} = \gamma\varepsilon_{ijl}$ with

$$\gamma = -(2/\epsilon_0\omega^2)\overline{\alpha}^{\rm gme} \tag{3.86}$$

according to Eq. (3.84). The standard expression[54, 36] $\rho = (\omega^2/2c^2)\mathrm{Re}\,\gamma$ for the rotatory strength becomes $\rho = -(1/\epsilon_0 c^2)\,\mathrm{Re}\,\overline{\alpha}^{\rm gme}$. Together with Eq. (11), this implies that the optical activity of a clean conductor goes to a constant at low frequencies. Instead, the optical activity of an insulator decreases as ω^2, because γ goes to a constant, as follows from Eq. (3.84) and the ω^2 scaling at low frequencies of $\mathrm{Im}\,\Pi^{\rm A}_{ijl}(\omega) = \omega\mathrm{Re}\,\sigma^{\rm A}_{ijl}(\omega)$: see, e.g., Ref. [48].

As an example, consider a Weyl semimetal with isotropic Weyl nodes described by the Hamiltonian (see main text)

$$H_{\mathbf{k}\nu} = \epsilon_\nu \mathbb{1} + \chi_\nu \hbar v_{\rm F}\mathbf{k}\cdot\boldsymbol{\sigma}\,. \tag{3.87}$$

The orbital contribution to $\overline{\alpha}^{\rm gme}$ from a pair of nodes of opposite chirality ($\nu = \mathrm{L}, \mathrm{R}$) is given in the clean semiclassical limit by the prefactor in Eq. (17), yielding

$$\boxed{\rho = (2\alpha/3hc)(\epsilon_{\rm L} - \epsilon_{\rm R})} \tag{3.88}$$

per node pair in the frequency range $1/\tau \ll \omega \ll \epsilon_{\rm gap}/\hbar$, where $\alpha = (1/4\pi\epsilon_0)(e^2/\hbar c) \approx 1/137$ is the fine-structure constant. This result, given as Eq. (18) in the main text, can be extended to lower frequencies (in the relaxation-time approximation) by inserting a factor of $\mathrm{Re}\,[i\omega\tau/(i\omega\tau - 1)] = \omega^2\tau^2/(1+\omega^2\tau^2)$ on the right-hand-side. Therefore ρ decreases as ω^2 in dirty metals, the same low-frequency behavior as in molecules[9] and insulators.[54, 48]

3.5.5.2 Nonequilibrium optical gyrotropy in Weyl semimetals driven by the chiral anomaly

It was pointed out in Ref. [35] that the chiral-anomaly mechanism leads to circular dichroism for light propagating inside a Weyl semimetal in the presence of electric and magnetic fields $\mathbf{E}_0$ and $\mathbf{B}_0$ with $\mathbf{E}_0\cdot\mathbf{B}_0 \neq 0$. In the following we use our microscopic formulation of natural gyrotropy in metals to calculate the induced response at low frequencies.

For Weyl nodes described by Eq. (3.87), we obtain from Eqs. (11) and (15) the following expression for the orbital contribution to $\alpha^{\rm gme}_{ij}(\omega) = \overline{\alpha}^{\rm gme}(\omega)\delta_{ij}$ from the spherical pocket

enclosing the ν-th node, with chemical potential μ_ν,

$$\overline{\alpha}_\nu^{\rm gme}(\omega) = \frac{1}{3}\frac{e}{(2\pi)^2 h}\frac{i\omega\tau}{i\omega\tau-1}(4\pi k_{\rm F}^2)\left(\mp\chi_\nu\frac{ev_{\rm F}}{2k_{\rm F}}\right) \tag{3.89}$$

$$= \mp\frac{1}{3}\frac{e^2}{h^2}\frac{i\omega\tau}{i\omega\tau-1}\chi_\nu\hbar v_{\rm F}k_{\rm F} = \frac{1}{3}\frac{e^2}{h^2}\frac{i\omega\tau}{i\omega\tau-1}\chi_\nu(\epsilon_\nu-\mu_\nu)\,. \tag{3.90}$$

The minus (plus) sign in the intermediate expressions corresponds to $\epsilon_\nu < \mu_\nu$ ($\epsilon_\nu > \mu_\nu$).

Consider a minimal model where $\nu = {\rm R}, {\rm L}$. In the equilibrium situation ($\mu_{\rm L} = \mu_{\rm R} = \epsilon_{\rm F}$) depicted in Fig. 1(b) in the main text, Eq. (3.89) reduces to Eq. (16) for $\omega\tau \gg 1$. Here we are interested in the scenario where the background $\mathbf{E}_0 \cdot \mathbf{B}_0$ field pumps charge across the nodes, leading to $\mu_{\rm R} \neq \mu_{\rm L}$. Following Ref.[35] we assume $\omega\tau \ll 1$, and for simplicity we set $\epsilon_{\rm L} = \epsilon_{\rm R} \equiv \epsilon_{\rm W}$ as in Fig. 1(a), so that the equilibrium gyrotropic response of Eq. (3.88) vanishes. Combining Eqs. (3.86) and (3.89) and taking the imaginary part yields

$$\mathrm{Im}\,\gamma_\nu(\omega \ll 1/\tau) = -\frac{\chi_\nu(\mu_\nu-\epsilon_{\rm W})e^2\tau}{6\pi^2\epsilon_0\hbar^2\omega}\,. \tag{3.91}$$

Summing over WPs and using $\sum_\nu \chi_\nu = 0$[57] gives for the orbital contribution to the nonequilibrium circular dichroism

$$\mathrm{Im}\,\gamma(\omega \ll 1/\tau) = \frac{(\mu_{\rm L}-\mu_{\rm R})\,e^2\tau}{6\pi^2\epsilon_0\hbar^2\omega}\,. \tag{3.92}$$

Equation (3.91) agrees with Eq. (11) of Ref. [35], except possibly for the numerical prefactor. Exact agreement was not expected, since the calculation in Ref. [35] was based on an incomplete formulation of optical gyrotropy in metals in terms of the Berry curvature. On the other hand, semiquantitative agreement for two-band models seems plausible in view of the simple relation between the Berry curvature and the orbital moment in such models.[82]

Consider now the nonequilibrium optical rotation. According to Eq. (3.89) $\mathrm{Re}\,\gamma(\omega \ll 1/\tau) = 0$, suggesting an absence of optical rotation at the lowest frequencies as found in Ref. [35]. Note, however, that Eq. (3.89) was obtained starting from Eq. (3.63), which assumes an equilibrium situation where the current response vanishes in the static limit according to Eqs. (3.60) and (3.66). In the presence of a chemical-potential imbalance a new reactive term appears in the gyrotropic response: the optical **B**-field induces an extra current at $\mathcal{O}(q)$ via Eq. (3), leading to optical rotation even for $\omega \ll 1/\tau$. Interestingly, this is a genuine Berry-curvature – as opposed to orbital-moment – contribution to (nonequilibrium) optical gyrotropy.

3.5.5.3 Reciprocity relation for a metal with a smooth interface

There is a general reciprocity principle for electromagnetic fields interacting with time-reversal (T) invariant media in equilibrium that requires the optical rotation in the reflection geometry to vanish.[32, 24] Thus, while for a T-breaking material there are in general optical

rotation effects both in transmission (Faraday) and in reflection (Kerr), optical rotation in T-invariant materials occurs in transmission only. At first glance this is surprising, because it implies a constraint on the nonlocal response functions imposed by T symmetry when the gyrotropic coefficient varies in space, for example at an interface.[3, 14, 72] This has been actively discussed recently in the context of optical rotation measured in reflection on cuprate superconductors.[36, 5, 24]

We show in the following how this constraint appears in our treatment of natural optical activity in metals. While the validity of the constraint is not in question, i.e., it had to be satisfied, it seems worthwhile to explain how it arises, given the recent interest in the nonlocal constitutive relation in spatially varying media with natural optical activity.

A T-invariant spatially-dispersive medium with a boundary or an interface is described, taking into consideration only the first derivatives with respect to coordinates, by the following constitutive relation,

$$D_i(\omega,\mathbf{r}) = \epsilon_0 \left[\epsilon^{\rm S}_{ij}(\omega,\mathbf{r})E_j(\omega,\mathbf{r}) + \gamma^{\rm A}_{ijl}(\omega,\mathbf{r})\partial_{r_l}E_j(\omega,\mathbf{r}) + E_j(\omega,\mathbf{r})\partial_{r_l}\lambda^{\rm A}_{ijl}(\omega,\mathbf{r})\right] . \tag{3.93}$$

This is Eq. (8) in Ref. [3], with the T-breaking parameter $\mathbf{B}_{\rm ext}$ set to zero. Under those conditions, the response tensors satisfy $\epsilon^{\rm S}_{ij}(\omega,\mathbf{r}) = \epsilon^{\rm S}_{ji}(\omega,\mathbf{r})$, $\gamma^{\rm A}_{ijl}(\omega,\mathbf{r}) = -\gamma^{\rm A}_{jil}(\omega,\mathbf{r})$, and $\lambda^{\rm A}_{ijl}(\omega,\mathbf{r}) = -\lambda^{\rm A}_{jil}(\omega,\mathbf{r})$. (More generally, in a T-breaking material the constitutive relation can be split into even and odd parts with respect to $\mathbf{B}_{\rm ext}$, and Eq. (3.93) then corresponds to the T-even part.) For an infinite macroscopically homogeneous medium the response tensors are independent of $\mathbf{r}$, and only the first two terms survive in Eq. (3.93). The last term is an additional contribution coming from the spatial inhomogeneity of the medium near the interface. The reciprocity constraint takes the form[3]

$$\lambda^{\rm A}_{ijl}(\omega,\mathbf{r}) = \frac{1}{2}\gamma^{\rm A}_{ijl}(\omega,\mathbf{r}) , \tag{3.94}$$

which in turn implies vanishing optical rotation in reflection: see Ref. [24] and references cited therein.

To obtain Eq. (3.94) for a metal subject to a low-frequency optical field, recall from the discussion around Eq. (7) that the natural gyrotropy of metals at low frequencies is fully determined by the magnetoelectric response of the medium, i.e., by the dynamic polarization $\mathbf{P}^{\mathbf{B}}$ and magnetization $\mathbf{M}^{\mathbf{E}}$ induced by the optical fields $\mathbf{B}$ and $\mathbf{E}$ respectively. In the main text, the T-even magnetoelectric response of a metal was calculated at $\hbar\omega \ll \epsilon_{\rm gap}$ using a semiclassical Boltzmann formalism, and the result was of the form

$$P^{\mathbf{B}}_i(\omega,\mathbf{r}) = (i/\omega)\alpha^{\rm gme}_{ij}(\omega,\mathbf{r})B_j(\omega,\mathbf{r}) \tag{3.95a}$$

$$M^{\mathbf{E}}_i(\omega,\mathbf{r}) = -(i/\omega)\alpha^{\rm gme}_{ji}(\omega,\mathbf{r})E_j(\omega,\mathbf{r}) , \tag{3.95b}$$

with $\boldsymbol{\alpha}^{\rm gme}$ given in the long-wavelength limit by Eq. (11). (The calculation was done for a bulk metal where $\boldsymbol{\alpha}^{\rm gme}$ is independent of $\mathbf{r}$, but the semiclassical approach remains valid if

we assume a smooth interface with a spatial variation that is slow on the scale of the mean free path.) Writing the constitutive relation in terms of the auxiliary fields,

$$\mathbf{D}(\omega,\mathbf{r}) = \epsilon_0\mathbf{E}(\omega,\mathbf{r}) + \mathbf{P}(\omega,\mathbf{r}) + (i/\omega)\boldsymbol{\partial}_{\mathbf{r}}\times\mathbf{M}(\omega,\mathbf{r})\,, \tag{3.96}$$

and using Eq. (3.95) together with $B_j = -(i/\omega)\varepsilon_{jlp}\partial_{r_l}E_p$ we find for the spatially-dispersive part of $D_i(\omega,\mathbf{r})$

$$\underbrace{(1/\omega^2)\left[\varepsilon_{ilp}\alpha^{\rm gme}_{jp}(\omega,\mathbf{r}) - \varepsilon_{jlp}\alpha^{\rm gme}_{ip}(\omega,\mathbf{r})\right]}_{\epsilon_0\gamma^{\rm A}_{ijl}(\omega,\mathbf{r})}\partial_{r_l}E_j(\omega,\mathbf{r}) + E_j(\omega,\mathbf{r})\partial_{r_l}\underbrace{\left[(1/\omega^2)\varepsilon_{ilp}\alpha^{\rm gme}_{jp}(\omega,\mathbf{r})\right]}_{\epsilon_0\lambda_{ijl}(\omega,\mathbf{r})}\,. \tag{3.97}$$

We have identified the tensors $\gamma^{\rm A}_{ijl}$ and $\lambda_{ijl} = \lambda^{\rm S}_{ijl} + \lambda^{\rm A}_{ijl}$ by comparing with Eq. (3.93), noting that the term containing $\lambda^{\rm S}_{ijl} = \lambda^{\rm S}_{jil}$ was included in the first term of Eq. (3.93).[3] The expression for $\gamma^{\rm A}_{ijl}$ is consistent with Eqs. (3.84) and (5a) for a bulk medium, and by inspection we obtain Eq. (3.94), with $2\lambda^{\rm A}_{ijl}(\omega,\mathbf{r}) \equiv \lambda_{ijl}(\omega,\mathbf{r}) - \lambda_{jil}(\omega,\mathbf{r})$. In conclusion, our microscopic formulation of natural gyrotropy in metals is consistent with the general reciprocity principle.

3.5.5.4 Berry-curvature contributions

In Ref. [86] the natural optical activity of clean metals was studied using a semiclassical "Berry-Boltzmann" approach, and a combination of the band velocity and the Berry curvature was shown to give a traceless contribution to the gyration tensor. Here we show how that contribution appears as part of the full microscopic expression in terms of the intrinsic magnetic moment. We start by setting $\omega\tau \gg 1$ in Eq. (9) to get

$$\alpha^{\rm gme}_{ij} = -e\sum_n\int[d\mathbf{k}]\,(\partial f/\partial\epsilon_{\mathbf{k}n})v_{\mathbf{k}n,i}\left(m^{\rm spin}_{\mathbf{k}n,j} + m^{\rm orb}_{\mathbf{k}n,j}\right) = \alpha^{\rm spin}_{ij} + \alpha^{\rm orb}_{ij}\,, \tag{3.98}$$

and note that Eq. (10) for $\mathbf{m}^{\rm orb}_{\mathbf{k}n}$ contains two terms, one of which involves the Berry curvature,

$$\mathbf{m}^{\rm orb}_{\mathbf{k}n} = (e/2\hbar){\rm Im}\langle\boldsymbol{\partial}_{\mathbf{k}}u_{\mathbf{k}n}|\times H_{\mathbf{k}}|\boldsymbol{\partial}_{\mathbf{k}}u_{\mathbf{k}n}\rangle + (e/2\hbar)\epsilon_{\mathbf{k}n}\boldsymbol{\Omega}_{\mathbf{k}n} = \mathbf{m}^{\rm H}_{\mathbf{k}n} + \mathbf{m}^{\Omega}_{\mathbf{k}n}\,. \tag{3.99}$$

Accordingly we write $\boldsymbol{\alpha}^{\rm orb} = \boldsymbol{\alpha}^{\rm H} + \boldsymbol{\alpha}^{\Omega}/2$, with

$$\alpha^{\rm H}_{ij} = -\frac{e^2}{2\hbar}\varepsilon_{jlp}\sum_n\int[d\mathbf{k}]\,(\partial f/\partial\epsilon_{\mathbf{k}n})v_{\mathbf{k}n,i}{\rm Im}\langle\partial_l u_{\mathbf{k}n}|H_{\mathbf{k}}|\partial_p u_{\mathbf{k}n}\rangle \tag{3.100a}$$

$$\alpha^{\Omega}_{ij} = -\frac{e^2}{\hbar}\sum_n\int[d\mathbf{k}]\,(\partial f/\partial\epsilon_{\mathbf{k}n})v_{\mathbf{k}n,i}\epsilon_{\mathbf{k}n}\Omega_{\mathbf{k}n,j}\,. \tag{3.100b}$$

The reason for writing $\boldsymbol{\alpha}^{\Omega}/2$ is that $\boldsymbol{\alpha}^{\rm H}$ can be further decomposed as (see derivation at the end)

$$\alpha^{\rm H}_{ij} = (1/2)\alpha^{\Omega}_{ij} + \Delta\alpha^{\rm orb}_{ij} \tag{3.101a}$$

$$\Delta\alpha_{ij}^{\rm orb} = \frac{e^2}{2\hbar^2}\varepsilon_{jlp}\sum_{n,m}\int [d\mathbf{k}]\, f^0_{\mathbf{k}n}(1-f^0_{\mathbf{k}m})\partial_i\left[(\epsilon_{\mathbf{k}n}+\epsilon_{\mathbf{k}m})\right.$$
$$\left.\mathrm{Im}(\langle\partial_l u_{\mathbf{k}n}|u_{\mathbf{k}m}\rangle\langle u_{\mathbf{k}m}|\partial_p u_{\mathbf{k}n}\rangle)\right]\,, \tag{3.101b}$$

so that finally

$$\boldsymbol{\alpha}^{\rm orb} = \boldsymbol{\alpha}^{\Omega} + \Delta\boldsymbol{\alpha}^{\rm orb}\,. \tag{3.102}$$

The decomposition (3.102) has two noteworthy features. Firstly, the Berry-curvature contribution is traceless,

$$\mathrm{tr}\left(\boldsymbol{\alpha}^{\Omega}\right) = \frac{e^2\epsilon_{\rm F}}{2\hbar^2}\sum_{n,a}\int_{S_{na}} dS\,(\hat{\mathbf{v}}_{\rm F}\cdot\boldsymbol{\Omega}_{\mathbf{k}n}) = \frac{\pi e^2\epsilon_{\rm F}}{\hbar^2}\sum_{n,a} C_{na} = 0\,, \tag{3.103}$$

for the same reason that the chiral magnetic effect vanishes when all Fermi sheets are in chemical equilibrium (see main text). Secondly, $\boldsymbol{\alpha}^{\Omega}$ and $\Delta\boldsymbol{\alpha}^{\rm orb}$ are separately gauge-invariant, but they are *not* separately invariant under a shift of the zero of energy. Nevertheless, $\boldsymbol{\alpha}^{\Omega}$ can be further decomposed into two traceless parts, only one of which depends on the zero of energy, by replacing $v_{\mathbf{k}n,i}\partial f/\partial\epsilon_{\mathbf{k}n}$ in Eq. (3.100b) with $(1/\hbar)\partial_i f^0_{\mathbf{k}n}$ and integrating by parts,

$$\alpha_{ij}^{\Omega} = \alpha_{ij}^{\Omega,1} + \alpha_{ij}^{\Omega,2} \tag{3.104a}$$
$$\alpha_{ij}^{\Omega,1} = \frac{e^2}{\hbar^2}\sum_n\int [d\mathbf{k}]\, f^0_{\mathbf{k}n}\epsilon_{\mathbf{k}n}\partial_i\Omega_{\mathbf{k}n,j} \tag{3.104b}$$
$$\alpha_{ij}^{\Omega,2} = \frac{e^2}{\hbar}\sum_n\int [d\mathbf{k}]\, f^0_{\mathbf{k}n}v_{\mathbf{k}n,i}\Omega_{\mathbf{k}n,j}\,. \tag{3.104c}$$

The trace of $\boldsymbol{\alpha}^{\Omega,1}$ vanishes for the same reason as Eq. (3.79), and the trace of $\boldsymbol{\alpha}^{\Omega,2}$ vanishes according to Eq. (2). Clearly $\boldsymbol{\alpha}^{\Omega,1}$ depends on the zero of energy, but $\boldsymbol{\alpha}^{\Omega,2}$ does not.

The Berry-curvature contribution identified in Ref. [86] amounts to $\boldsymbol{\alpha}^{\Omega,2}$. To see this, convert Eq. (3.104c) into a (traceless) contribution to the gyration tensor using Eq. (3.85),

$$g_{ij}^{\Omega,2} = \frac{e^2}{\hbar\omega c\epsilon_0}\sum_n\int [d\mathbf{k}]\, f^0_{\mathbf{k}n}v_{\mathbf{k}n,j}\Omega_{\mathbf{k}n,i}\,. \tag{3.105}$$

To compare with Ref. [86] the expression above should be multiplied by $ic\epsilon_0$,[8] leading to Eq. (10) therein.[9]

In closing, we reiterate that even if Berry-curvature contributions can be identified (see also Sec. 3.5.4), the magnetic moment on the FS provides a more natural and compact description of the low-frequency natural gyrotropy of metals.

[8] In Ref. [86] the gyration tensor $\tilde{\mathbf{g}}$ was introduced via the relation $j_i^{\rm g} = -i\varepsilon_{ijm}\tilde{g}_{ml}E_j q_l$, where $\boldsymbol{j}^{\rm g}$ is the natural gyrotropy current $j_i^{\rm g} = \Pi^{\rm A}_{ijl}A_j q_l$. Using Eqs. (3.81a) and (3.84) we find $j_i^{\rm g} = c\epsilon_0\varepsilon_{ijm}g_{ml}E_j q_l$, where $\mathbf{g}$ is the gyration tensor as defined in this work. Therefore, $\tilde{g}_{ij} = ic\epsilon_0 g_{ij}$.

[9] The sign convention for both the Berry curvature and for Fourier transforms is the opposite in Ref. [86] compared to the present work, but this does not affect the combination $(1/\omega)\Omega_{\mathbf{k}n,i}$ appearing in Eq. (3.105).

Derivation of Eq. (3.101) Integrating Eq. (3.100a) by parts and inserting a complete set of states gives, in the condensed notation used earlier,

$$\begin{aligned}\alpha_{ij}^{\rm H} &= \frac{e^2}{2\hbar^2}\varepsilon_{jlp}\sum_{n,m}\int [d\mathbf{k}]\ f_n^0\ \partial_i\left[\epsilon_m {\rm Im}(\langle\partial_l n|m\rangle\langle m|\partial_p n\rangle)\right] \\ &= \frac{e^2}{2\hbar^2}\varepsilon_{jlp}\sum_{n,m}\int [d\mathbf{k}]\ \left[f_m^0 - f_m^0\left(1-f_n^0\right) + f_n^0\left(1-f_m^0\right)\right]\partial_i\left[\epsilon_m {\rm Im}(\langle\partial_l n|m\rangle\langle m|\partial_p n\rangle)\right]\ .\end{aligned}$$

Exchanging n and m in the first and second terms,

$$\begin{aligned}\alpha_{ij}^{\rm H} &= \frac{e^2}{2\hbar^2}\varepsilon_{jlp}\sum_{n,m}\int [d\mathbf{k}]\ \Big\{f_n^0\ \partial_i\left[\epsilon_n {\rm Im}(\langle\partial_l m|n\rangle\langle n|\partial_p m\rangle)\right] \\ &- f_n^0\left(1-f_m^0\right)\partial_i\left[\epsilon_n {\rm Im}(\langle\partial_l m|n\rangle\langle n|\partial_p m\rangle)\right] \\ &+ f_n^0\left(1-f_m^0\right)\partial_i\left[\epsilon_m {\rm Im}(\langle\partial_l n|m\rangle\langle m|\partial_p n\rangle)\right]\Big\}\,. \qquad (3.106)\end{aligned}$$

Using Eq. (3.41) to combine the second and the third terms and to recast the first in terms of the Berry curvature we obtain, after an integration by parts,

$$\begin{aligned}\alpha_{ij}^{\rm H} = &- \frac{e^2}{2\hbar}\sum_n\int [d\mathbf{k}]\ \left(\partial f/\partial\epsilon_n\right) v_{n,i}\left(\epsilon_n\Omega_{n,j}\right) \\ &+ \frac{e^2}{2\hbar^2}\varepsilon_{jlp}\sum_{n,m}\int [d\mathbf{k}]\ f_n^0\left(1-f_m^0\right)\partial_i\left[(\epsilon_n+\epsilon_m){\rm Im}(\langle\partial_l n|m\rangle\langle m|\partial_p n\rangle)\right]\ , \qquad (3.107)\end{aligned}$$

which is Eq. (3.101).

Chapter 4

Magnetoresistance and nonlinear optical responses in metals

We study nonlinear optical responses of metals using a Floquet approach. We show that the semiclassical formula for the circular photogalvanic effect arising from the Berry curvature dipole is reproduced by a full quantum calculation using a Floquet approach. We derive formulas for the magnetoconductivity by a semiclassical approach including both the Berry curvature and orbital magnetic moment. Applied to Weyl fermions, the semiclassical approach captures the directional anisotropy of linear conductivity under magnetic field as a consequence of an anisotropic B^2 contribution, which may explain the low-field regime of recent experiments.

4.1 Introduction

The wavefunction of a single electron moving through a crystal has several geometric properties whose importance in insulators is well known. The most celebrated example is the Berry phase derived from Bloch states. It gives a gauge field in momentum space that underlies topological phases ranging from the integer quantum Hall effect to topological insulators. These phases are characterized by topological invariants that can be expressed as integrals of Berry gauge fields; even in ordinary insulators, similar integrals describe important physical quantities such as electric polarization [68, 42] as well as the magnetoelectric response [63, 22, 49].

In metals, the Berry gauge field is known to give an additional term (the "anomalous velocity") in the semiclassical equations of motion that describe the motion in real and momentum space of a wavepacket made from Bloch states. The anomalous velocity was originally discussed in the context of the anomalous Hall effect in magnetic metals such as iron. The semiclassical equations can be derived systematically to linear order in applied electric and magnetic fields, under certain assumptions that we review more fully in Sec. 4.2 below. In several cases, such as the anomalous Hall effect [53] and the gyrotropic or "trans-

port limit" of the chiral magnetic effect [85, 47], the semiclassical approach (SCA) fully reproduces the results obtained from quantum-mechanical calculations based on the Kubo formula.

The focus of this chapter is the semiclassical theory of *nonlinear* properties of metals that are currently active subjects of experimental and theoretical investigation. One motivation is that systematic quantum-mechanical derivations that capture all contributions to a given nonlinear order in applied fields have not as yet been achieved. An example we consider is the chiral anomaly, which in a solid is a particular type of angle-dependent magnetoresistance with an enhanced electrical conductivity along the direction of an applied magnetic field. This effect has been argued to exist based on linearization near isolated Dirac or Weyl singularities, but the lesson of the past few years of work on the chiral magnetic effect is that it can be dangerous to treat the singularities solely and without including all effects at a given order. We derive a semiclassical formula for magnetotransport in the weak-field regime of this problem, and discuss that including all terms gives an answer distinct from that in other recent work, which may explain experimental observations on a Dirac semimetal in this regime [46, 78].

The semiclassical equations of motion for an electron wavepacket in a metal are [67]

$$\dot{\boldsymbol{r}} = \frac{1}{\hbar}\boldsymbol{\nabla}_{\boldsymbol{k}}\epsilon_{\boldsymbol{k}} - \dot{\boldsymbol{k}} \times \boldsymbol{\Omega}, \tag{4.1a}$$

$$\hbar\dot{\boldsymbol{k}} = -e\boldsymbol{E} - e\dot{\boldsymbol{r}} \times \boldsymbol{B}. \tag{4.1b}$$

One new contribution compared to the version in older textbooks [7] is from the Berry curvature in momentum space,

$$\Omega = -\mathrm{Im}[\langle\nabla_{\boldsymbol{k}}u_{\boldsymbol{k}}| \times |\nabla_{\boldsymbol{k}}u_{\boldsymbol{k}}\rangle], \tag{4.2}$$

and another is from the orbital magnetic moment contribution to the energy dispersion: $\epsilon_{\boldsymbol{k}} = \epsilon^0_{\boldsymbol{k}} - \boldsymbol{m}_{\boldsymbol{k}} \cdot \boldsymbol{B}$ where $H_{\boldsymbol{k}}|u_{\boldsymbol{k}}\rangle = \epsilon^0_{\boldsymbol{k}}|u_{\boldsymbol{k}}\rangle$ with $B = 0$ and the orbital magnetic moment is

$$\boldsymbol{m}_{\boldsymbol{k}} = -\frac{e}{2\hbar}\mathrm{Im}[\langle\nabla_{\boldsymbol{k}}u_{\boldsymbol{k}}| \times (H_{\boldsymbol{k}} - \epsilon^0_{\boldsymbol{k}})|\nabla_{\boldsymbol{k}}u_{\boldsymbol{k}}\rangle]. \tag{4.3}$$

(We note that we adopt the convention $e > 0$.)

The SCA equations conserve the properly defined volume in phase space and give an intuitive approach to many observable properties of metals. However, the SCA can make erroneous predictions if used outside the regime of its validity. To illustrate this point we present, in Sec. 4.2, the predictions of semiclassical and fully quantum theories of a fundamental nonlinear response in metals with low symmetry - the photogalvanic effect (PGE) [25, 20, 60]. The term "photogalvanic" refers to the generation of a dc current by a time-varying electric field, with amplitude proportional to the square of the applied field. The PGE is distinguished from a conventional photovoltaic response by the dependence of the dc current on the polarization state of the electric field. For example, in the the circular PGE (CPGE) the direction of the dc current reverses when the polarization state of the

time-varying field is changed from left to right circular. Using the SCA the CPGE has been shown to have a Berry-phase contribution [61] in 2D and more recently in 3D [64] systems such as Weyl semimetals.

In Sec. 4.2 we show that the previous semiclassical predictions for the CPGE can be derived in a fully quantum theory by using the Floquet approach [52]. We first derive the Berry curvature formula for CPGE in the case of two band and then generalize the derivation to the cases with many bands. This indicates that the CPGE provides a good example where the nonlinear effects that follow from semiclassical equations are exactly what is obtained from a full quantum derivation, which was previously only known in the linear case. We also show that in this same limit in which interband terms are neglected, there is close quantitative relation between CPGE and second-harmonic generation (SHG).

In Sec. 4.3 we derive semiclassical formulas for magento conductance. In Sec. 4.4, we apply our semiclassical formula to magneto-transport of Weyl/Dirac semimetals and study the angle-dependent magnetoresistance. We find that there exist contributions of opposite sign from orbital magnetic moment and Berry curvature in addition to the contribution of the chiral anomaly. The angular dependence that we obtain by taking into account all the contributions at the same order in the SCA is compared with recent magnetotransport experiments [46, 78].

4.2 Nonlinear optical effects and Floquet approach

In this section, we first review formulas for the nonlinear Kerr rotation and CPGE. Previous works based on SCA showed that those nonlinear optical effects are described by a geometrical quantity, i.e., Berry curvature dipole [64]. We give an alternative derivation for those formulas based on fully quantum theoretical treatment by applying Floquet formalism for a two-band system.

4.2.1 Geometrical meaning of nonlinear optics in the semiclassical approach

In previous semiclassical works [61, 64], it has been shown that the intraband contributions to SHG and CPGE have a geometrical nature that are described by Berry curvatures of Bloch wave functions. The SHG is the second order nonlinear optical effect that is described by nonlinear current responses $\boldsymbol{j}(2\omega)e^{-2i\omega t}$ as

$$j_a^{(2\omega)} = \sigma_{abc} E_b E_c, \tag{4.4}$$

when the external electric field is given by

$$\boldsymbol{E}(t) = \boldsymbol{E}e^{-i\omega t} + \boldsymbol{E}^* e^{i\omega t}. \tag{4.5}$$

Nonlinear Hall effect in Ref. [64] refers to a transverse current response that is described by σ_{abb} with $a \neq b$. Similarly, the CPGE is the second order nonlinear optical effect in which dc photocurrent of $\boldsymbol{j}^{(0)}$ is induced by circularly polarized light as

$$j_a^{(0)} = \sigma_{abc} E_b E_c^*. \tag{4.6}$$

In a time reversal symmetric material, these nonlinear response tensors σ are given by

$$\sigma_{abc} = \epsilon_{adc} \frac{e^3 \tau}{\hbar(1 - i\omega\tau)} \int [d\mathbf{k}] f_0 (\partial_b \Omega_d), \tag{4.7}$$

when the frequency ω is much smaller than the resonant frequency for optical transitions (i.e., the intraband contribution). Here, ϵ_{abc} is the totally antisymmetric tensor, f_0 is the Fermi distribution function, and we used the notation $[d\mathbf{k}] = d\boldsymbol{k}/(2\pi)^d$ with the dimension d.

We focus here on the case of a 3D material [64] but have adopted slightly different notations for $\boldsymbol{E}(t)$ and j from those in Ref. [64], which resulted in a modified expression for σ above. While these nonlinear effects are Fermi surface effects because one obtains $\sigma_{abc} \propto \epsilon_{adc} \int [d\mathbf{k}] (\partial_b f_0) \Omega_d$ by integrating by parts, they can be also understood as currents carried by electrons in the Fermi sea with anomalous velocity originating from the Berry curvature dipole.

The way that the anomalous velocity ($\dot{\boldsymbol{k}} \times \boldsymbol{\Omega}$) of electron wave packets driven by an external electric field leads to CPGE and SHG is schematically illustrated in Fig. 4.1. Circular polarized light induces circular motion of the wave packet in momentum space [Fig. 4.1(a)]. In the Berry curvature dipole, the anomalous velocities in regions with $\Omega > 0$ and $\Omega < 0$ add, which results in dc current. Similarly, linearly polarized light induces an oscillation of wave packet as shown in Fig. 4.1(b). The driven wave packet exhibits anomalous velocities in the y direction that oscillate twice in the driving period, which results in SHG.

4.2.2 Fully quantum mechanical derivation by Floquet formalism

We study the nonlinear optical effects including CPGE and SHG from a fully quantum mechanical treatment by using a two band model. The focus of interest is whether the fully quantum mechanical expression coincides with the semiclassical formula. While SCA partially includes high energy bands through Ω, it does not necessarily capture all effects of the high energy bands. Thus it is an interesting question whether the geometrical formulas for CPGE and SHG hold even in the fully quantum mechanical treatment. In the following, we study the intraband contribution to CPGE and SHG by applying the Floquet formalism and show that the Berry curvature formulas is indeed exact in the fully quantum mechanical treatment.

First we study a two band system periodically driven by an external electric field by using the Floquet formalism (for details of Floquet formalism, see Refs. [43, 59, 52]). When

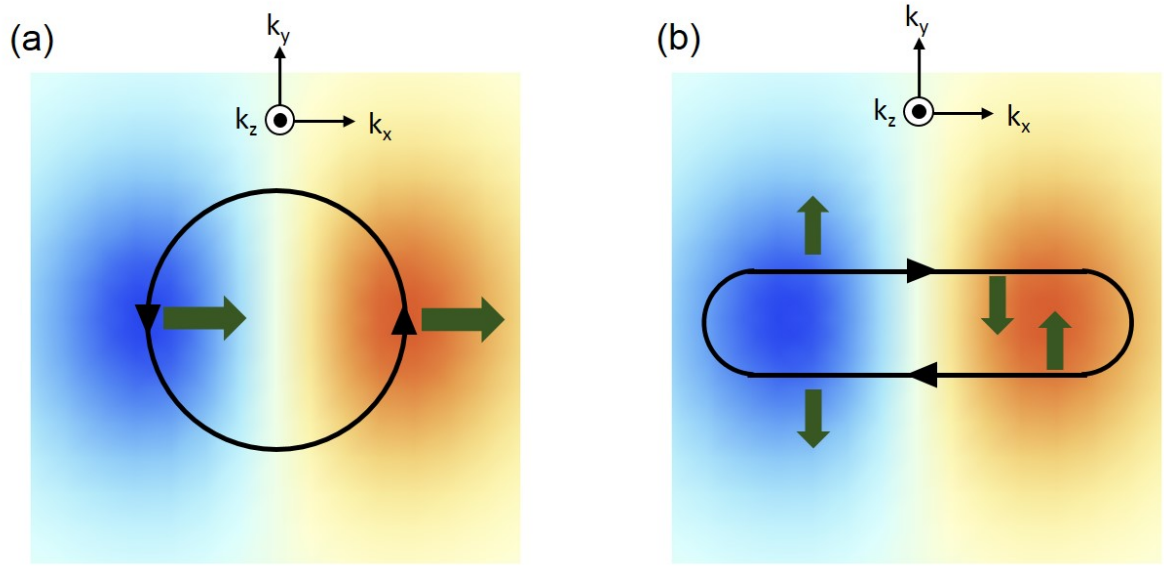

Figure 4.1: Semiclassical picture of CPGE and SHG induced by a Berry curvature dipole. The distribution of Berry curvature in momentum space is indicated by the color scale, with red region corresponding to $\Omega_z > 0$ and blue region to $\Omega_z < 0$. (a) CPGE arises from circular motion of the electron wave packet in momentum space driven by circularly polarized light. The dipole structure in $\Omega(k)$ induces an anomalous velocity ($\dot{\boldsymbol{k}} \times \boldsymbol{\Omega}$) in the x direction denoted by green arrows. (b) SHG arises from oscillation of electron wave packet driven by linearly polarized light in the x direction. The Berry curvature dipole leads to an anomalous velocity that undergoes two oscillations in the y direction in one driving period. The shown configuration of Berry curvature preserves C_{2v} point group symmetry (which is present for typical polar crystals that support CPGE and SHG), where the y-axis corresponds to the polar axis and the yz-plane to the mirror plane.

the original Hamiltonian of the two band system is given by a Bloch Hamiltonian $H_{\text{orig}}(k)$, the time dependent Hamiltonian of the system driven by $E(t) = Ee^{-i\omega t} + E^* e^{i\omega t}$ is given by

$$H(t,k) = H_{\text{orig}}(k + eA(t)), \tag{4.8}$$

$$A(t) = i\frac{E}{\omega}e^{-i\omega t} - i\frac{E^*}{\omega}e^{i\omega t}, \tag{4.9}$$

which is periodic in time with $t \to t + 2\pi/\omega$. For such periodically driven systems, the Floquet formalism gives a concise description in terms of band picture as follows. The Floquet formalism is, roughly speaking, a time-direction analog of Bloch's theorem for time-dependent Hamiltonian $H(t)$ that satisfies $H(t+T) = H(t)$ with period T. Namely, in a

similar manner to Bloch's theorem, the solution for the time-periodic Schrödinger equation,

$$i\hbar\frac{\partial\psi(t)}{\partial t} = H(t)\psi(t), \tag{4.10}$$

is given by a time-periodic form

$$\psi(t) = e^{-i\epsilon t/\hbar}\phi(t), \qquad \phi(t+T) = \phi(t), \tag{4.11}$$

with the quasienergy ϵ. By using the time-periodic part of the wave function $\phi(t)$, the time-dependent Schrödinger equation is rewritten as

$$(i\hbar\partial_t + \epsilon)\phi(t) = H(t)\phi(t). \tag{4.12}$$

Since $\phi(t)$ is periodic in time, we can perform Fourier transformation of the both sides with

$$\phi(t) = \sum_m e^{-im\omega t}\phi_m, \tag{4.13}$$

and obtain

$$(m\hbar\omega + \epsilon)\phi_m = \widetilde{H}_{mn}\phi_n, \tag{4.14}$$

$$\widetilde{H}_{mn} = \frac{1}{T}\int_0^T dt e^{i(m-n)\omega t}H(t). \tag{4.15}$$

Here $\widetilde{H}_{mn}$ is time-independent, but has an additional matrix structure spanned by Floquet indices m and n. Thus the time-dependent Schrödinger equation effectively reduces to a time-independent one in the Floquet formalism as,

$$H_F\phi = \epsilon\phi, \tag{4.16}$$

where the Floquet Hamiltonian is given by

$$(H_F)_{mn} = \frac{1}{T}\int_0^T dt e^{i(m-n)\omega t}H(t) - n\hbar\omega\delta_{mn}. \tag{4.17}$$

Floquet bands obtained by diagonalizing the Floquet Hamiltonian H_F offer a concise understanding of the dynamics of a driven system in terms of an effective band picture. We note that the energy spectrum of ϵ shows a periodic structure with $\hbar\omega$ as a consequence of translation symmetry with respect to the Floquet index n. Thus the quasienergy spectrum is essentially described within the range $-\hbar\omega/2 \leq \epsilon < \hbar\omega/2$, which is an analog of "the first Brillouin zone" in Bloch's theorem.

Since we consider the case of driving frequency much lower than the band gap, we can obtain the current expectation value by studying the Floquet band that is connected to

the valence band in the undriven system. In order to do so, we use standard second order perturbation theory for

$$H_F = H_0 + H_1 + H_2, \tag{4.18}$$

where H_i represents a term in the Floquet Hamiltonian proportional to A^i. The wave function up to the second order in A reads

$$\begin{aligned} |\psi_n\rangle = |n\rangle &- \sum_{m\neq n} \frac{(H_1)_{mn}}{E_m - E_n}|m\rangle \\ &\sum_{m\neq n}\Bigg[- \frac{(H_2)_{mn}}{E_m - E_n} - \frac{(H_1)_{mn}(H_1)_{nn}}{(E_m - E_n)^2} \\ &+ \sum_{k\neq n} \frac{(H_1)_{mk}(H_1)_{kn}}{(E_m - E_n)(E_k - E_n)}\Bigg]|m\rangle, \end{aligned} \tag{4.19}$$

where $H_0|n\rangle = E_n|n\rangle$. By applying the above formula to the Floquet Hamiltonian H_F, we obtain Floquet states $|\psi\rangle$ that describes the steady state under the drive of incident light. The current responses in the steady state are obtained from perturbed Floquet states that are connected to the original valence bands. This treatment can be justified when the frequency of incident light is much smaller than the energy difference of valence and conduction bands. (When ω satisfies conditions for optical resonances, Floquet bands originating from valence and conduction bands anticross each other. In this case, we cannot naively determine occupation of resulting Floquet bands, which requires considering the coupling to a heat bath [52].)

By using the Floquet state $|\psi\rangle$ connected to the valence band, the time dependent current in the steady state is given by

$$J_\alpha(t) = \sum_{m,n} \{\mathrm{tr}[|\psi\rangle\langle\psi|\hat{v_\alpha}]\}_{mn} e^{-i(m-n)\omega t}, \tag{4.20}$$

where tr denotes the trace over the band index, m, n are Floquet indices, and $\hat{v_\alpha}$ is the current operator along the α-direction is given by

$$(\hat{v_\alpha})_{mn} = \frac{1}{T}\int_0^T dt e^{i(m-n)\omega t}\frac{\partial H(t)}{\partial k_\alpha}. \tag{4.21}$$

In the following, we derive representative components of nonlinear response tensor describing CPGE and SHG by using the above method.

To study CPGE we consider a system subjected to the left circularly polarized light in the xy plane, where the electric field is given by

$$\boldsymbol{E}(t) = E(\boldsymbol{e_x} + i\boldsymbol{e_y})e^{-i\omega t} + E^*(\boldsymbol{e_x} - i\boldsymbol{e_y})e^{i\omega t}. \tag{4.22}$$

In this case, the Floquet Hamiltonian is written as

$$H_F = H_0 + H_1, \tag{4.23}$$

$$(H_0)_{mn} = \begin{pmatrix} \epsilon_v - n\omega & 0 \\ 0 & \epsilon_c - n\omega \end{pmatrix} \delta_{mn}, \tag{4.24}$$

$$(H_1)_{mn} = -iA^*(v_x - iv_y)\delta_{mn-1} + iA(v_x + iv_y)\delta_{mn+1}, \tag{4.25}$$

where $\epsilon_{v/c}$ is energy of valence/conduction band, $v_i = \partial H_0/\partial k_i$ is the velocity operator for the static Hamiltonian, $A = E/\omega$, and we set $e = 1, \hbar = 1$ for simplicity. Here we dropped the term H_2 proportional to A^2 because it does not contribute to dc photocurrent which is proportional to AA^* and does not involve A^2 terms in the end. Since we are interested in the second order nonlinear current responses, it is sufficient to consider the Floquet Hamiltonian with $n = -2, \ldots, 2$ by starting with the unperturbed wave function $|\psi_{ini}\rangle = |u_{v,n=0}\rangle$. Now we study dc current in the x-direction induced by circularly polarized light for the steady state described by the Floquet state in Eq. (4.19). The velocity operator in the x-direction is written up to linear order in A as

$$\begin{aligned} \hat{v_x} = v_x\delta_{mn} &- iA^*\partial_{k_x}(v_x - iv_y)\delta_{mn-1} \\ &+ iA\partial_{k_x}(v_x + iv_y)\delta_{mn+1}. \end{aligned} \tag{4.26}$$

By using Eq. (4.20), we obtain the CPGE photocurrent $J_x = \int [d\mathbf{k}] j_x^{(0)}$ as

$$\begin{aligned} j_x^{(0)} &= \sum_n \{\mathrm{tr}[|\psi\rangle\langle\psi|\hat{v_x}]\}_{nn} \\ &= 4\frac{|E|^2}{\omega}\Bigg\{ \frac{\mathrm{Im}[(\partial_{k_x} v_x)_{vc}(v_y)_{cv} + (v_x)_{vc}(\partial_{k_x} v_y)_{cv}]}{(\epsilon_v - \epsilon_c)^2} \\ &\qquad - 3\frac{\mathrm{Im}[(v_x)_{vc}(v_y)_{cv}][(v_x)_{vv} - (v_x)_{cc}]}{(\epsilon_v - \epsilon_c)^3} \Bigg\}, \end{aligned} \tag{4.27}$$

where we dropped higher order terms with respect to ω by focusing on the current response in the low frequency limit. We note that the contributions proportional to $|E|^2/\omega^2$ vanish due to the time reversal symmetry (e.g., the TRS $T = \mathcal{K}$ constrains $\mathrm{Re}[v]$ and $\mathrm{Im}[v]$ to be odd and even functions of k, respectively), which is used when going from the first line to the second line. In the case of two band models, the Berry curvature is written as

$$\Omega_z = -\frac{2\mathrm{Im}[(v_x)_{vc}(v_y)_{cv}]}{(\epsilon_v - \epsilon_c)^2}, \tag{4.28}$$

and the matrix elements of $\partial_{k_i} v_j$ can be rewritten as

$$(\partial_{k_i} v_j)_{vc} = \partial_{k_i}[(v_j)_{vc}] + (v_j)_{vc}\,[i(a_i)_v - i(a_i)_c]$$

$$+ (v_i)_{vc}\frac{(v_j)_{vv} - (v_j)_{cc}}{\epsilon_v - \epsilon_c}, \tag{4.29}$$

with $(a_i)_{v/c} = \langle u_{v/c}|\partial_{k_i}|u_{v/c}\rangle$. By using these formulas, the CPGE photocurrent can be further reduced as

$$\begin{aligned} j_x^{(0)} &= 4\frac{|E|^2}{\omega}\frac{\partial}{\partial k_x}\left[\frac{\mathrm{Im}[(v_x)_{vc}(v_y)_{cv}]}{(\epsilon_v - \epsilon_c)^2}\right] \\ &= -2\frac{|E|^2}{\omega}\partial_{k_x}\Omega_z. \end{aligned} \tag{4.30}$$

The nonlinear conductivity tensor is obtained by equating the above expression and j_x in terms of σ and $\boldsymbol{E}(t)$ [in Eq. (4.22)] given by

$$j_x = -i\sigma_{xxy}|E|^2 + i\sigma_{xyx}|E|^2 = -2i\sigma_{xxy}|E|^2. \tag{4.31}$$

Here we used antisymmetry of imaginary part of σ with respect to the last two indices. This leads to

$$\sigma_{xxy} = \frac{1}{i\omega}\int [d\mathbf{k}]\partial_{k_x}\Omega_z, \tag{4.32}$$

and reproduces the semiclassical formula for σ_{xxy} in Eq. (4.7). We note that the factor $\tau/(1-i\omega\tau)$ in the semiclassical formula [Eq. (4.7)] is replaced by the factor i/ω in the above formula because the $\tau \to \infty$ limit (clean limit) is effectively taken in the Floquet perturbation theory.

Next we study SHG by using Floquet perturbation theory and the two band model in a similar manner to CPGE. We consider a system driven by linearly polarized light in the x direction as $E_x(t) = Ee^{-i\omega t} + E^* e^{i\omega t}$ and the SHG in the y direction. The corresponding Floquet Hamiltonian is given by

$$H_F = H_0 + H_1 + H_2, \tag{4.33}$$

$$(H_0)_{mn} = \begin{pmatrix} \epsilon_v - n\omega & 0 \\ 0 & \epsilon_c - n\omega \end{pmatrix}\delta_{mn}, \tag{4.34}$$

$$(H_1)_{mn} = \left(-iA^*\delta_{mn-1} + iA\delta_{mn+1}\right)v_x, \tag{4.35}$$

$$(H_2)_{mn} = \left(-\frac{(A^*)^2}{2}\delta_{mn-2} + |A|^2\delta_{mn} - \frac{A^2}{2}\delta_{mn+2}\right)\partial_{k_x}v_x. \tag{4.36}$$

We take $|\psi_{ini}\rangle = |u_{v,n=0}\rangle$ as the unperturbed wave function and keep the part of the Floquet Hamiltonian within the range $n = -2, \ldots, 2$. The velocity operator along the y-direction is given by

$$\begin{aligned} \hat{v_y} = v_y\delta_{mn} &+ \left(-iA^*\delta_{mn-1} + iA\delta_{mn+1}\right)\partial_{k_x}v_y \\ &+ \left(-\frac{(A^*)^2}{2}\delta_{mn-2} + |A|^2\delta_{mn} - \frac{A^2}{2}\delta_{mn+2}\right)\partial_{k_x}^2 v_y. \end{aligned} \tag{4.37}$$

By using Eq. (4.20), we obtain the Fourier component of the current $J_y = \int [d\mathbf{k}] j_y$ proportional to $e^{-2i\omega t}$ as

$$\begin{aligned} j_y^{(2\omega)} &= \sum_n \{\mathrm{tr}[|\psi\rangle\langle\psi|\hat{v_y}]\}_{n+2,n} \\ &= -2i\frac{E^2}{i\omega}\frac{\partial}{\partial k_x}\left[\frac{\mathrm{Im}[(v_x)_{vc}(v_y)_{cv}]}{(\epsilon_v-\epsilon_c)^2}\right] \\ &= i\frac{E^2}{\omega}\partial_{k_x}\Omega_z. \end{aligned} \tag{4.38}$$

Here we again used the fact that the contributions proportional to E^2/ω^2 vanish due to the time reversal symmetry, and also dropped contributions with higher powers of ω. The above expression indicates that the nonlinear conductivity tensor σ_{yxx} is written as

$$\sigma_{yxx} = \frac{i}{\omega}\int [d\mathbf{k}]\partial_{k_x}\Omega_z. \tag{4.39}$$

This again reproduces the semiclassical formula for σ_{yxx} in Eq. (4.7).

We can extend the above analysis based on the Floquet formalism to general cases with many bands and obtain the same Berry curvature dipole formula. We sketch the derivation in the following (for details, see supplemental materials 4.5). We consider the general Floquet Hamiltonian under the light irradiation which is given by

$$H_F = H_0 + H_1 + H_2, \tag{4.40}$$

with

$$H_1 = \sum_i A_i v_i, \qquad H_2 = \frac{1}{2}\sum_{i,j} A_i A_j \partial_{k_i} v_j, \tag{4.41}$$

where H_0 represents a static Hamiltonian with many bands. By using the Floquet perturbation theory in Eq. (4.19) and the expression for the current in Eq. (4.20), we obtain the general expression for the nonlinear current response as

$$\begin{aligned} J_r = &-\sum_{i,j} A_i A_j \int [d\mathbf{k}] \\ &\times\Bigg[\sum_{n,g}[f(\epsilon_n)-f(\epsilon_g)]\left(\frac{1}{2}\frac{(v_r)_{ng}(\partial_{k_i}v_j)_{gn}}{\epsilon_n-(\epsilon_g+2\omega)}+\frac{(\partial_r v_j)_{ng}(v_i)_{gn}}{\epsilon_n-(\epsilon_g+\omega)}\right) \\ &+\sum_{n,g,m}\left(\frac{f(\epsilon_n)}{\epsilon_n-\epsilon_m-\omega}-\frac{f(\epsilon_g)}{\epsilon_g-\epsilon_m+\omega}\right)\frac{(v_r)_{ng}(v_i)_{gm}(v_j)_{mn}}{\epsilon_n-(\epsilon_g+2\omega)} \\ &+\sum_{n,g,m} f(\epsilon_n)\frac{(v_j)_{nm}(v_r)_{mg}(v_i)_{gn}}{(\epsilon_n-(\epsilon_g+\omega))(\epsilon_n-(\epsilon_m-\omega))} \\ &+\sum_n \frac{1}{2}f(\epsilon_n)(\partial_{k_r}\partial_{k_i}v_j)_{nn}\Bigg], \end{aligned} \tag{4.42}$$

with Fermi distribution function $f(\epsilon)$ [where $f(\epsilon_n) = 1(0)$ for occupied (unoccupied) states]. When we expand the current J_r with respect to ω, the lowest order contribution in ω is proportional to ωA^2 in the presence of time reversal symmetry. In the case of many bands, the Berry curvature dipole for the nth band is written as

$$\partial_{k_i}\Omega_{z,n} = -2\mathrm{Im}\left[\frac{\langle n|\partial_{k_x}H|m\rangle\langle m|\partial_{k_y}H|n\rangle}{(\epsilon_n-\epsilon_m)^2}\right], \tag{4.43}$$

where n runs over occupied bands and m runs over unoccupied bands. By using this expression for the Berry curvature dipole, it turns out that the lowest order contribution of J_y proportional to ωA^2 is written as

$$J_y = -iwA_x^2\int[d\mathbf{k}]f(\epsilon_n)\partial_{k_x}\Omega_{z,n}, \tag{4.44}$$

which reproduces the Berry curvature dipole formula Eq. (4.7) for SHG in the case of many bands. Details of the above calculation for many band cases are described in Appendix 4.5.

To summarize, we derived formulas for CPGE and SHG in the sufficiently low frequency region in a fully quantum mechanical way by using Floquet perturbation theory. This reproduces the semiclassical formula with Berry curvature dipole.

4.3 Semiclassical formulas for magnetoconductance

We apply SCA to obtain semiclassical formula for nonlinear magneto-conductivity that includes all terms proportional to B^2E is applicable to Weyl semimetals and may explain directional anisotropy of magnetoconductivity of Weyl semimetals recently reported in Ref. [46, 78], which we perform in Sec. 4.4. Third, TR symmetric Weyl semimetals can support large nonlinear Kerr rotation. Intraband contribution to SHG vanishes for $B = 0$ in TR symmetric Weyl semimetals, and the SHG signal has a contribution linear in B. Thus application of B may lead to giant nonlinear Kerr rotation.

We derive semiclassical formulas for nonlinear magneto-optical effects up to the second order in E. It is convenient to rewrite the equations of motion (4.1) to collect time derivatives on the left:

$$\dot{\boldsymbol{r}} = \frac{1}{\hbar D}[\boldsymbol{\nabla}_{\boldsymbol{k}}\epsilon_{\boldsymbol{k}} + e\boldsymbol{E}\times\boldsymbol{\Omega}_{\boldsymbol{k}} + \frac{e}{\hbar}(\boldsymbol{\nabla}_{\boldsymbol{k}}\epsilon_{\boldsymbol{k}}\cdot\boldsymbol{\Omega}_{\boldsymbol{k}})\boldsymbol{B}], \tag{4.45}$$

$$\hbar\dot{\boldsymbol{k}} = \frac{1}{D}[-e\boldsymbol{E} - \frac{e}{\hbar}\boldsymbol{\nabla}_{\boldsymbol{k}}\epsilon_{\boldsymbol{k}}\times\boldsymbol{B} - \frac{e^2}{\hbar}(\boldsymbol{E}\cdot\boldsymbol{B})\boldsymbol{\Omega}_{\boldsymbol{k}}], \tag{4.46}$$

$$D = 1 + \frac{e}{\hbar}\boldsymbol{B}\cdot\boldsymbol{\Omega}_{\boldsymbol{k}}. \tag{4.47}$$

The charge density ρ and current density $\boldsymbol{j}$ are given by

$$\rho = -e\int[d\boldsymbol{k}]Df, \tag{4.48}$$

$$\boldsymbol{j} = -e \int [d\boldsymbol{k}](D\dot{\boldsymbol{r}} + \boldsymbol{\nabla}_{\boldsymbol{r}} \times \boldsymbol{m}_{\boldsymbol{k}})f, \tag{4.49}$$

with $[d\boldsymbol{k}] = d\boldsymbol{k}/(2\pi)^3$, where the second term of $\boldsymbol{j}$ is a contribution of magnetization current. We note that the factor D arises from a field-induced change of the volume of the phase space [77]. In the following, we focus on the uniform system. In this case, the expression of the current density reduces to

$$\boldsymbol{j} = -e \int [d\boldsymbol{k}][\tilde{\boldsymbol{v}}_{\boldsymbol{k}} + \frac{e}{\hbar}\boldsymbol{E} \times \boldsymbol{\Omega}_{\boldsymbol{k}} + \frac{e}{\hbar}(\tilde{\boldsymbol{v}}_{\boldsymbol{k}} \cdot \boldsymbol{\Omega}_{\boldsymbol{k}})\boldsymbol{B}]f, \tag{4.50}$$

where we used

$$\tilde{\boldsymbol{v}}_{\boldsymbol{p}} = \boldsymbol{v}_{\boldsymbol{k}} - (1/\hbar)\boldsymbol{\nabla}_{\boldsymbol{k}}(\boldsymbol{m} \cdot \boldsymbol{B}), \tag{4.51}$$

with $\boldsymbol{v}_{\boldsymbol{k}} = (1/\hbar)\boldsymbol{\nabla}_{\boldsymbol{k}}\epsilon^0_{\boldsymbol{k}}$.

Now we focus on responses driven by monochromatic light with the electric field $\boldsymbol{E}(t) = \boldsymbol{E}e^{-i\omega t}$. We consider current responses at order of E. We write the distribution function in Fourier components as

$$f = f_0 + f_1 e^{-i\omega t}, \tag{4.52}$$

where f_0 is the unperturbed distribution function and other terms appear in the presence of the electric field of the incident light. The steady-state distribution function is determined by the Boltzmann equation

$$\frac{df}{dt} = \frac{f_0 - f}{\tau}, \tag{4.53}$$

where

$$\frac{df}{dt} = \dot{\boldsymbol{k}} \cdot \boldsymbol{\nabla}_{\boldsymbol{k}} f + \partial_t f. \tag{4.54}$$

In the following, we apply the above SCA to the linear current responses in the presence of magnetic fields.

We derive the semiclassical formula for the conductivity up to the second order of B in terms of Berry curvature and orbital magnetic moment.

The current response of the frequency ω is obtained from f_1 in Eq. (4.52). By equating terms proportional to $e^{-i\omega t}$ in Eq. (4.54), we obtain

$$[-e\boldsymbol{E} - \frac{e^2}{\hbar}(\boldsymbol{E} \cdot \boldsymbol{B})\boldsymbol{\Omega}_{\boldsymbol{k}}] \cdot \boldsymbol{\nabla}_{\boldsymbol{p}} f_0 - i\omega f_1 = -\frac{f_1}{\tau}, \tag{4.55}$$

with $\boldsymbol{\nabla}_{\boldsymbol{p}} = (1/\hbar)\boldsymbol{\nabla}_{\boldsymbol{k}}$, where we dropped the term involving $(\boldsymbol{\nabla}_{\boldsymbol{k}}\epsilon_{\boldsymbol{k}}) \times \boldsymbol{B}$ because it is perpendicular to $\boldsymbol{\nabla}_{\boldsymbol{p}} f_0 = (1/\hbar)(\boldsymbol{\nabla}_{\boldsymbol{k}}\epsilon_{\boldsymbol{k}})\partial_\epsilon f_0$. This leads to

$$f_1 = \frac{-\tau}{1 - i\omega\tau}\frac{1}{D}[-e\boldsymbol{E} - \frac{e^2}{\hbar}(\boldsymbol{E} \cdot \boldsymbol{B})\boldsymbol{\Omega}_{\boldsymbol{k}}] \cdot \boldsymbol{\nabla}_{\boldsymbol{p}} f_0. \tag{4.56}$$

Now the current response linear in E is given by

$$\boldsymbol{j_1} = \frac{e\tau}{1-i\omega\tau}\int_{\mathrm{BZ}}[d\boldsymbol{k}]\frac{1}{D}\Big\{[\tilde{\boldsymbol{v}}_{\boldsymbol{k}} + \frac{e}{\hbar}(\tilde{\boldsymbol{v}}_{\boldsymbol{k}}\cdot\boldsymbol{\Omega}_{\boldsymbol{k}})\boldsymbol{B}] \\ \times[-e\boldsymbol{E} - \frac{e^2}{\hbar}(\boldsymbol{E}\cdot\boldsymbol{B})\boldsymbol{\Omega}_{\boldsymbol{k}}]\cdot\boldsymbol{\nabla}_{\boldsymbol{p}}f_0 + \frac{e}{\hbar}\boldsymbol{E}\times\boldsymbol{\Omega}_{\boldsymbol{k}}f_0\Big\}, \tag{4.57}$$

where $f_0 = \theta(E_F - \epsilon_{\boldsymbol{k}} - \boldsymbol{m}_{\boldsymbol{k}}\cdot\boldsymbol{B})$ with the step function $\theta(x) = 0(x<0), 1(x\geq 0)$. This expression is reduced if we focus on the case where the electric field $\boldsymbol{E}$ is applied along the ith direction and the system preserves the TRS in the absence of magnetic fields. Specifically, we consider terms up to $\propto \boldsymbol{B}$ that are nonvanishing with the TRS by expanding as $1/D \simeq 1 + (e/\hbar)\boldsymbol{B}\cdot\boldsymbol{\Omega}_{\boldsymbol{k}}$, which leads to

$$\boldsymbol{j_1} = \frac{e\tau}{1-i\omega\tau}\int_{\mathrm{BZ}}[d\boldsymbol{k}]\{-\boldsymbol{v}_{\boldsymbol{k}}eE(\boldsymbol{v}_{\boldsymbol{k}})_i\partial_\epsilon f_0' \\ + \frac{e}{\hbar}(\boldsymbol{E}\times\boldsymbol{\Omega}_{\boldsymbol{k}})(\boldsymbol{m}\cdot\boldsymbol{B})\partial_\epsilon f_0'\}, \tag{4.58}$$

with $f_0' = \theta(E_F - \epsilon_{\boldsymbol{k}})$, i.e., a distribution function when $\boldsymbol{B} = 0$. Here we used the fact that $\partial_{p_i}, \boldsymbol{v}_{\boldsymbol{p}}, \boldsymbol{\Omega},$, and $\boldsymbol{m}$ are odd under the TRS. The first term in the integral is the metallic conductivity, while the second term describes regular Hall conductivity linear in B (in contrast to anomalous Hall conductivity which is nonzero in the absence of B). This second term indicates that the orbital magnetic moment gives a semiclassic description related to Landau level formation in the quantum limit. We note that there is no B-linear contribution to the longitudinal conductivity σ_{ii} because the Onsager relation constrains the conductivity as $\sigma_{ij}(B) = \sigma_{ji}(-B)$ and the longitudinal conductivity should be an even function of B.

Next, we derive the formula for the longitudinal magnetoconductance. Its lowest order dependence on B is quadratic due to the Onsager relation. The B^2 contribution to the longitudinal current response is explicitly written as

$$\boldsymbol{j}_{B^2} = \frac{e^2\tau}{\hbar}\int_{\mathrm{BZ}}[d\boldsymbol{k}]\Big\{ -\frac{e}{\hbar}\mathbf{E}\cdot\boldsymbol{\nabla}_{\boldsymbol{k}}f_0(\epsilon^0)[-e(\boldsymbol{v}_{\boldsymbol{k}}\cdot\boldsymbol{\Omega}_{\mathbf{k}})(\mathbf{B}\cdot\boldsymbol{\Omega}_{\mathbf{k}})\mathbf{B} - e\boldsymbol{\Omega}_{\mathbf{k}}\cdot\boldsymbol{\nabla}_{\boldsymbol{k}}(\boldsymbol{m}\cdot\mathbf{B})\mathbf{B} \\ + e(\mathbf{B}\cdot\boldsymbol{\Omega}_{\mathbf{k}})^2\boldsymbol{v}_{\boldsymbol{k}} + (\mathbf{B}\cdot\boldsymbol{\Omega}_{\mathbf{k}})\boldsymbol{\nabla}_{\boldsymbol{k}}(\boldsymbol{m}\cdot\mathbf{B})] + [\frac{1}{\hbar}\mathbf{E}\cdot\boldsymbol{\nabla}_{\boldsymbol{k}}(\frac{\partial f_0(\epsilon^0)}{\partial\epsilon}\boldsymbol{m}\cdot\mathbf{B}) \\ - \frac{e}{\hbar}(\mathbf{E}\cdot\mathbf{B})(\boldsymbol{\Omega}_{\mathbf{k}}\cdot\boldsymbol{\nabla}_{\boldsymbol{k}}f_0(\epsilon^0))][e(\boldsymbol{v}_{\boldsymbol{k}}\cdot\boldsymbol{\Omega}_{\mathbf{k}})\mathbf{B} - e(\mathbf{B}\cdot\boldsymbol{\Omega}_{\mathbf{k}})\boldsymbol{v}_{\boldsymbol{k}} - \boldsymbol{\partial}_{\boldsymbol{k}}(\boldsymbol{m}\cdot\mathbf{B})] \\ + \frac{e}{\hbar}(\mathbf{E}\cdot\mathbf{B})[\boldsymbol{\Omega}_{\mathbf{k}}\cdot\boldsymbol{\partial}_{\boldsymbol{k}}(\frac{\partial f_0(\epsilon^0)}{\partial\epsilon}\boldsymbol{m}\cdot\mathbf{B})]\boldsymbol{v}_{\boldsymbol{k}} - \frac{1}{2}\mathbf{E}\cdot\boldsymbol{\partial}_{\boldsymbol{k}}[\frac{\partial^2 f_0(\epsilon^0)}{\partial\epsilon^2}(\boldsymbol{m}\cdot\mathbf{B})^2]\boldsymbol{v}_{\boldsymbol{k}}\Big\}. \tag{4.59}$$

In addition to terms that contribute isotropically to the current density, there are several terms that contribute to the current density specifically along $\boldsymbol{B}$ which results in an anisotropic magnetoconductance if it is applied to Weyl semimetals as we discuss in Sec. 4.4.

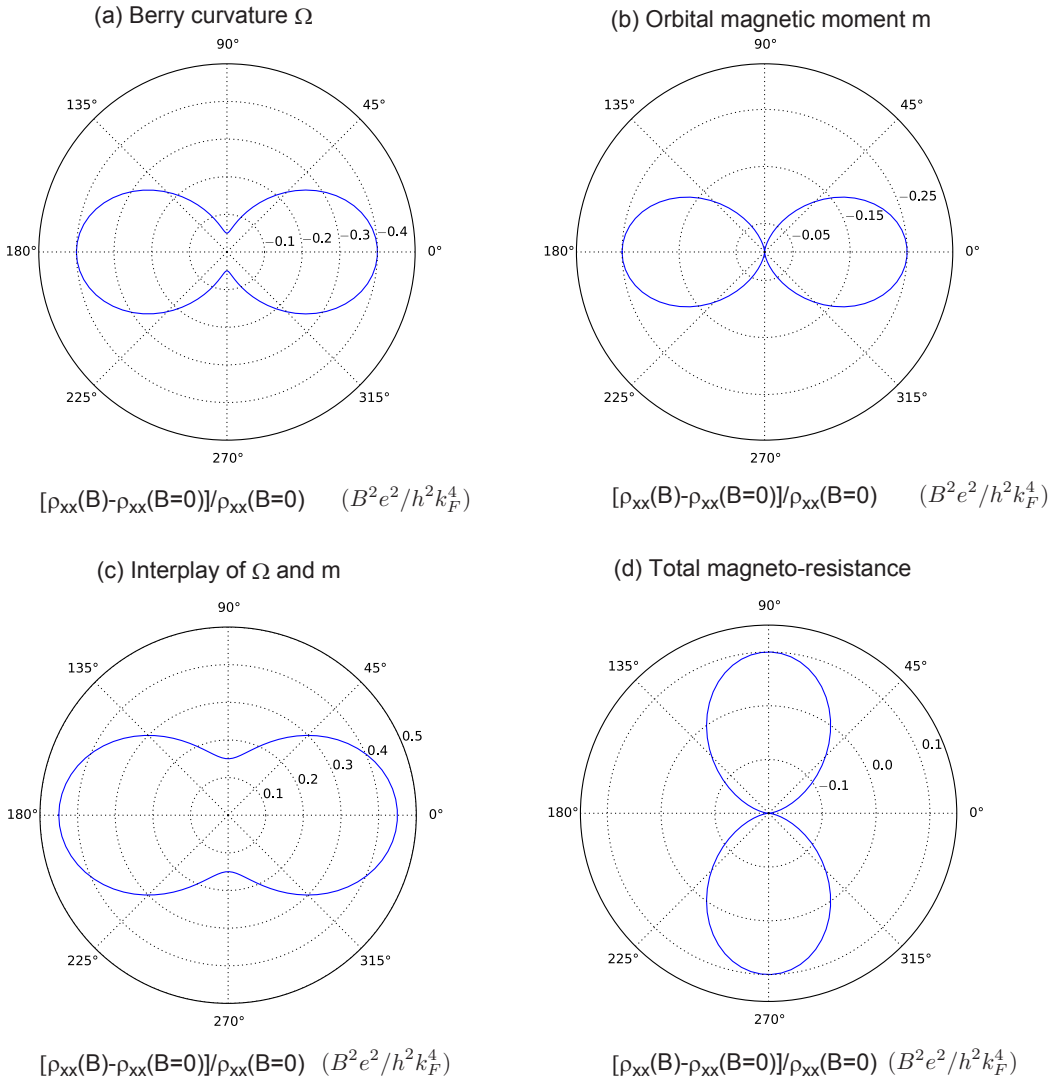

Figure 4.2: Angle-dependence of longitudinal magnetoresistance (LMR) for Weyl semimetals derived from the semiclassical approach [Eq. (4.59)]. Blue lines are polar plots of the LMR $[\rho_{xx}(B) - \rho_{xx}(B=0)]/\rho_{xx}(B=0)$ as a function of the relative angle θ between $\boldsymbol{E}$ and $\boldsymbol{B}$. We show angle-dependences of contributions to the LMR from (a) the Berry curvature, (b) the orbital magnetic moment, (c) the interplay between the Berry curvature and the orbital magnetic moment, and (d) the total angle-dependence of the LMR.

4.4 Angle-dependent magnetoresistance

In this section, we study magnetoresistance by using the SCA developed in the previous section. In particular, we focus on the current response $J \propto EB^2$ and study how the Berry curvature and the orbital magnetic moment contribute to magnetoresistance in Weyl semimetals, since the interplay of these two quantities in transport properties of Weyl semimetals has not been fully investigated except for a few studies [85, 47, 62, 70]. The obtained angle-dependence of magnetoresistance is compared with recent magneto-transport experiments for Dirac semimetals [46, 78].

We consider the Hamiltonian for Weyl semimetals given by

$$H = \eta v_F \boldsymbol{\sigma} \cdot \boldsymbol{p}, \tag{4.60}$$

where v_F is the Fermi velocity and $\eta = \pm 1$ specifies the chirality. In this case, the velocity operator, the Berry curvature, the orbital magnetic moment are written as

$$\boldsymbol{v_k} = v_F \hat{\boldsymbol{k}}, \tag{4.61}$$

$$\boldsymbol{\Omega} = -\eta \frac{1}{2k^2} \hat{\boldsymbol{k}}, \tag{4.62}$$

$$\boldsymbol{m} = -\eta \frac{e v_F}{2k} \hat{\boldsymbol{k}}, \tag{4.63}$$

for the conduction band, where $\hat{k}$ denotes the unit vector along $\boldsymbol{k}$.

Now we apply the semiclassics formula Eq. (4.59) for the linear current response $\boldsymbol{j_1}$ proportional to B^2 to Weyl semimetals and study the angle-dependent magnetoresistance. First, we suppose that the electric field is applied in the z-direction as $\boldsymbol{E} = E\boldsymbol{e_z}$ where $\boldsymbol{e_z}$ denotes the unit vector along the z direction. In this case, the current along the z-direction $(j_1)_z$ is given by

$$(j_1)_z = \frac{1}{6\pi^2\hbar} \tau e^2 v_F k_F^2 E + \frac{1}{30\pi^2\hbar^3 k_F^2} \tau e^4 v_F B^2 E \tag{4.64a}$$

when $\boldsymbol{E} \parallel \boldsymbol{B}$, and

$$(j_1)_z = \frac{1}{6\pi^2\hbar} \tau e^2 v_F k_F^2 E - \frac{1}{60\pi^2\hbar^3 k_F^2} \tau e^4 v_F B^2 E \tag{4.64b}$$

when $\boldsymbol{E} \perp \boldsymbol{B}$ (e.g. $\boldsymbol{B} \parallel \hat{x}$), where we assumed $\tau\omega \ll 1$. Here, the first term is the isotropic dc conductivity and the second term is an anisotropic correction which originates from the $\boldsymbol{E} \cdot \boldsymbol{B}$ term related to the chiral anomaly in Weyl semimetals. The second term accounts for the negative magnetoresistance (MR) when $\boldsymbol{E} \parallel \boldsymbol{B}$, and the positive MR when $\boldsymbol{E} \perp \boldsymbol{B}$. Thus the semiclassical theory for the linear conductivity including effects of both $\boldsymbol{\Omega}$ and $\boldsymbol{m}$ captures the directional anisotropy of linear conductivity in the $\boldsymbol{B}$ field which is usually considered to be an evidence of a Weyl fermion in transport measurements.

Next, we discuss the full angle dependence of the current response in the magnetic field. When the electric field is applied in the direction tilted by θ from the direction of the magnetic field $\boldsymbol{B}$, the longitudinal magneto conductivity $\sigma(B)$ is given by

$$\frac{\sigma(B)-\sigma(B=0)}{\sigma(B=0)} = \frac{-1+3\cos^2\theta}{10}\frac{e^2B^2}{\hbar^2 k_F^4}\,. \tag{4.65}$$

Equation (4.65) does not depend on the chirality of the Weyl node nor in which band the chemical potential is located. It shows that the magnetoresistance (MR) is positive when $\mathbf{E} \perp \mathbf{B}$ and it decreases to negative as $\theta \to 0$. If we separately look at contributions to the MR from the Berry curvature and the orbital magnetic moment, we find that either the Berry curvature or the orbital magnetic moment alone gives a negative magnetoresistance (Figs. 4.2a and 4.2b), while the interplay between the Berry curvature and the orbital magnetic moment gives a positive magnetoresistance (Fig. 4.2c). As a whole, Eq. (4.65) gives the angular dependences as shown in Fig. 4.2d. We note that the anisotropic magnetoconductance in the semiclassics [Eq. (4.65)] is not solely described by the contribution from the chiral anomaly. Specifically, the contribution from the chiral anomaly which was discussed in Ref. [65] is found in the term

$$\frac{-e^4\tau}{\hbar}\int_{\mathrm{BZ}}[d\boldsymbol{k}](\boldsymbol{\Omega}_{\mathbf{k}}\cdot\boldsymbol{\nabla}_{\boldsymbol{p}}f_0(\epsilon^0))(\boldsymbol{v}_{\boldsymbol{k}}\cdot\boldsymbol{\Omega}_{\mathbf{k}})(\mathbf{E}\cdot\mathbf{B})\mathbf{B} \tag{4.66}$$

in Eq. (4.59) and gives a negative magnetoresistance in Weyl semimetals. In contrast, there are several terms involving the orbital magnetic moment which lead to contributions of opposite signs.

A similar angular dependence of the magnetoresistance to Eq. (4.65) in the weak field region have been observed in magneto-transport experiments of Dirac semimetals [46, 78]. In particular, Ref. [78] reported that the sign change of the MR occurs around 45° in the low B region for Dirac semimetal Na_3Bi, which is consistent with our semiclassical result shown in Fig. 4.2(d). We note that our calculation for Weyl semimetals is also applicable to Dirac semimetals with a mild assumption that the degenerate energy bands having opposite chirality in Dirac semimetals are decoupled with each other.

Finally, we present estimates for the above nonlinear conductivities derived for Weyl semimetals. The directional anisotropy of the linear conductivity is given by the ratio of the two terms $\propto B^0$ and $\propto B^2$ in Eq. (4.64). The anisotropy ratio amounts to $0.06(B/1\ \mathrm{T})^2$ for typical parameters $v_F = 3\times10^5$ m/s, $E_F = 10$ meV for the Weyl semimetal material, TaAs [73, 39].

4.5 Supplemental material for derivation of the Berry curvature dipole formula for general bands

In this appendix, we apply the Floquet perturbation theory to systems with a general number of bands and derive the formula for SHG in terms of Berry curvature dipole. The derivation

proceeds in a similar manner to the two band case presented in Sec. 4.2B, but with involving more band indices.

We consider the system irradiated with monochromatic light which is described by the time-dependent Hamiltonian,

$$\begin{aligned}\widetilde{H}(t) &= H(\boldsymbol{p}+e\boldsymbol{A}(t)) = H^0 + H^1 + H^2 + \cdots \\ &= H + \sum_i (\partial_{k_i} H) e A_i e^{-i\omega t} + \sum_{i,j} \frac{1}{2}(\partial_{k_i}\partial_{k_j} H) e^2 A_i A_j e^{-2i\omega t} + \cdots ,\end{aligned} \tag{4.67}$$

where $H^0 \equiv H$ is the static Hamiltonian in the absence of the driving, and $\boldsymbol{A}(t) = \boldsymbol{A}e^{-i\omega t}$ is the vector potential. For the time periodic Hamiltonian $\widetilde{H}(t)$, the Floquet Hamiltonian is defined by

$$(H_F)_{mn} = \frac{1}{T}\int_0^T dt e^{i(m-n)\Omega t}\widetilde{H}(t) - n\hbar\Omega\delta_{mn}, \tag{4.68}$$

with Floquet indices m an n. In the following, we adopt a simplified notation where we write contributions $H^i(t)$ to the Floquet Hamiltonian H_F just by H^i.

The standard perturbation theory gives the wave function for the perturbed Floquet Hamiltonian H_F as

$$|\psi_{\tilde{n}}\rangle = |\tilde{n}\rangle + \sum_{\tilde{g}\neq\tilde{n}} \frac{H^1_{\tilde{g}\tilde{n}}}{\epsilon_{\tilde{n}} - \epsilon_{\tilde{g}}}|\tilde{g}\rangle + \sum_{\substack{\tilde{n}\neq\tilde{m}\\ \tilde{g}\neq\tilde{n}}} \left[\frac{H^1_{\tilde{g}\tilde{m}}H^1_{\tilde{m}\tilde{n}}}{(\epsilon_{\tilde{n}} - \epsilon_{\tilde{m}})(\epsilon_{\tilde{n}} - \epsilon_{\tilde{g}})} - \frac{H^1_{\tilde{n}\tilde{n}}H^1_{\tilde{g}\tilde{n}}}{(\epsilon_{\tilde{n}} - \epsilon_{\tilde{g}})^2} + \frac{H^2_{\tilde{g}\tilde{n}}}{\epsilon_{\tilde{n}} - \epsilon_{\tilde{g}}}\right]|\tilde{g}\rangle, \tag{4.69}$$

where $|\tilde{n}\rangle$ is the unperturbed wave function satisfying $H|\tilde{n}\rangle = \epsilon_{\tilde{n}}|\tilde{n}\rangle$, and $\tilde{n}$ labels the set of the band index and the Floquet index. Here we note that $H^1_{\tilde{n}\tilde{n}} = 0$ in the present case. The explicit form of the wave function ψ_n with the band index n and any Floquet index (say, 0) is given by

$$\begin{aligned}|\psi_n\rangle =& |n\rangle + e\sum_{n,g}\frac{(\partial_{k_i}HA_i)_{gn}}{\epsilon_n - (\epsilon_g+\omega)}|g\rangle + \frac{1}{2}e^2\sum_{n,g}\frac{(\partial_{k_i}\partial_{k_j}HA_iA_j)_{gn}}{\epsilon_n - (\epsilon_g+2\omega)}|g\rangle \\ &+ e^2\sum_{n,m,g}\left[\frac{(\partial_{k_j}HA_j)_{gm}(\partial_{k_i}HA_i)_{mn}}{(\epsilon_n - (\epsilon_m+\omega))(\epsilon_n - (\epsilon_g+2\omega))}\right]|g\rangle ,\end{aligned} \tag{4.70}$$

where $|n\rangle$ denotes the static wave function with the band index n, ϵ_n denotes the static energy dispersion with the band index n, and $\mathcal{O}_{m,n} = \langle m|\mathcal{O}|n\rangle$.

Now we consider the current response in the α-direction is given by

$$J_\alpha(t) = -e\sum_n f(\epsilon_n)\sum_{m',n'}\{\mathrm{tr}[|\psi_{(n,0)}\rangle\langle\psi_{(n,0)}|\hat{v_\alpha}]\}_{m'n'}e^{-i(m'-n')\omega t}, \tag{4.71}$$

where $|\psi_{(n,0)}\rangle$ is the perturbed wave function with the band index n and the Floquet index 0, and m', n' denote the Floquet indices. The Fermi distribution function $f(\epsilon)$ is given

by $f(\epsilon_n) = 1$ for occupied bands and $f(\epsilon_n) = 0$ for unoccupied bands. Since we consider the low-frequency limit where optical transition does not take place, we can assume that the occupation of the perturbed states coincides with that of the unperturbed states. The operator $\hat{v}$ is Floquet representation of the time-dependent velocity operator $v(t)$ which is given by

$$(\hat{v}_i)_{m'n'} = \frac{1}{T}\int_0^T dt e^{i(m'-n')\omega t} v_i(t) \tag{4.72}$$

$$\begin{aligned} v_i(t) &= v_i^0 + v_i^1 + v_i^2 + \cdots \\ &= \partial_{k_i} H + \sum_j (\partial_{k_i}\partial_{k_j} H) e A_j e^{-i\omega t} + \sum_{i,j} \frac{1}{2}(\partial_{k_i}\partial_{k_j}\partial_{k_l} H) e^2 A_j A_l e^{-2i\omega t} + \cdots . \end{aligned} \tag{4.73}$$

For the real external field $\boldsymbol{A}(t) = \boldsymbol{A}e^{-i\omega t} + \boldsymbol{A}e^{i\omega t}$, we obtain the second-order current response J_r along the r-direction which is proportional to $e^{-i2\omega t}$ as

$$\begin{aligned} J_r = -e^3 \sum_{i,j} A_i A_j \int [d\mathbf{k}] \sum_{n,g} \Bigg[& \frac{1}{2} f(\epsilon_n) \frac{(\partial_{k_i}\partial_{k_j} H)_{gn}}{\epsilon_n - (\epsilon_g + 2\omega)} \langle n|\partial_{k_r} H|g\rangle \\ & + \frac{1}{2} f(\epsilon_n) \frac{(\partial_{k_i}\partial_{k_j} H)_{ng}}{\epsilon_n - (\epsilon_g - 2\omega)} \langle g|\partial_{k_r} H|n\rangle \\ & + \sum_m \frac{f(\epsilon_n)}{\epsilon_n - \epsilon_m - \omega} \frac{(\partial_{k_j} H)_{gm}(\partial_{k_i} H)_{mn}}{\epsilon_n - (\epsilon_g + 2\omega)} \langle n|\partial_{k_r} H|g\rangle \\ & + \sum_m \frac{f(\epsilon_n)}{\epsilon_n - \epsilon_m + \omega} \frac{(\partial_{k_i} H)_{nm}(\partial_{k_j} H)_{mg}}{\epsilon_n - (\epsilon_g - 2\omega)} \langle g|\partial_{k_r} H|n\rangle \\ & + f(\epsilon_n) \frac{(\partial_{k_i} H)_{gn}}{\epsilon_n - (\epsilon_g + \omega)} \langle n|\partial_{k_r}\partial_{k_j} H|g\rangle + \ f(\epsilon_n) \frac{(\partial_{k_i} H)_{ng}}{\epsilon_n - (\epsilon_g - \omega)} \langle g|\partial_{k_r}\partial_{k_j} H|n\rangle \\ & + \sum_m f(\epsilon_n) \frac{(\partial_{k_i} H)_{gn}}{\epsilon_n - (\epsilon_g + \omega)} \frac{(\partial_{k_j} H)_{nm}}{\epsilon_n - (\epsilon_m - \omega)} \langle m|\partial_{k_r} H|g\rangle + \ \frac{1}{2} f(\epsilon_n) \langle n|\partial_{k_r}\partial_{k_i}\partial_{k_j} H|n\rangle \Bigg] . \end{aligned} \tag{4.74}$$

This expression can be rewritten as

$$\begin{aligned} J_r = -e^3 \sum_{i,j} A_i A_j \int [d\mathbf{k}] \sum_n \Bigg[& \sum_g \frac{1}{2}(f(\epsilon_n) - f(\epsilon_g)) \frac{\langle n|\partial_{k_r} H|g\rangle\langle g|\partial_{k_i}\partial_{k_j} H|n\rangle}{\epsilon_n - (\epsilon_g + 2\omega)} \\ & + \sum_{m,g} \Big(\frac{f(\epsilon_n)}{\epsilon_n - \epsilon_m - \omega} - \frac{f(\epsilon_g)}{\epsilon_g - \epsilon_m + \omega} \Big) \frac{\langle n|\partial_{k_r} H|g\rangle\langle g|\partial_{k_i} H|m\rangle\langle m|\partial_{k_j} H|n\rangle}{\epsilon_n - (\epsilon_g + 2\omega)} \\ & + \sum_g (f(\epsilon_n) - f(\epsilon_g)) \frac{\langle n|\partial_{k_r}\partial_{k_j} H|g\rangle\langle g|\partial_{k_i} H|n\rangle}{\epsilon_n - (\epsilon_g + \omega)} \\ & + \sum_{m,g} f(\epsilon_n) \frac{\langle n|\partial_{k_j} H|m\rangle\langle m|\partial_{k_r} H|g\rangle\langle g|\partial_{k_i} H|n\rangle}{(\epsilon_n - (\epsilon_g + \omega))(\epsilon_n - (\epsilon_m - \omega))} + \frac{1}{2} f(\epsilon_n) \langle n|\partial_{k_r}\partial_{k_i}\partial_{k_j} H|n\rangle \Bigg] . \end{aligned}$$

(4.75)

Since we are interested in the intraband effects in the low frequency limit (ω much smaller than the bandgap), we expand the current J_r in terms of ω as $J_r = J_r^0 + J_r^1 + J_r^2 + \ldots$, with $J_r^n \propto \omega^n$. The lowest order term in ω is the zeroth order term which is given by

$$\begin{aligned}
J_r^0 = -e^3 \sum_{i,j} A_i A_j \int [d\mathbf{k}] \sum_n \Bigg[& \sum_g \frac{1}{2}(f(\epsilon_n) - f(\epsilon_g)) \frac{\langle n|\partial_{k_r} H|g\rangle\langle g|\partial_{k_i}\partial_{k_j} H|n\rangle}{\epsilon_n - \epsilon_g} \\
& + \sum_{m,g}{}' \Big(\frac{f(\epsilon_n)}{\epsilon_n - \epsilon_m} - \frac{f(\epsilon_g)}{\epsilon_g - \epsilon_m} \Big) \frac{\langle n|\partial_{k_r} H|g\rangle\langle g|\partial_{k_i} H|m\rangle\langle m|\partial_{k_j} H|n\rangle}{\epsilon_n - \epsilon_g} \\
& + \sum_{g}{}' (f(\epsilon_n) - f(\epsilon_g)) \frac{\langle n|\partial_{k_r}\partial_{k_j} H|g\rangle\langle g|\partial_{k_i} H|n\rangle}{\epsilon_n - \epsilon_g} \\
& + \sum_{m,g}{}' f(\epsilon_n) \frac{\langle n|\partial_{k_j} H|m\rangle\langle m|\partial_{k_r} H|g\rangle\langle g|\partial_{k_i} H|n\rangle}{(\epsilon_n - \epsilon_g)(\epsilon_n - \epsilon_m)} \\
& - 2\sum_{g}{}' f(\epsilon_n) \frac{\langle n|\partial_{k_r} H|g\rangle\langle g|\partial_{k_i} H|n\rangle\langle n|\partial_{k_j} H|n\rangle}{(\epsilon_n - \epsilon_g)^2} \\
& - 2\sum_{g}{}' f(\epsilon_g) \frac{\langle n|\partial_{k_r} H|g\rangle\langle g|\partial_{k_i} H|g\rangle\langle g|\partial_{k_j} H|n\rangle}{(\epsilon_n - \epsilon_g)^2} \\
& + \sum_{m}{}' f(\epsilon_n) \frac{\langle n|\partial_{k_j} H|m\rangle\langle m|\partial_{k_r} H|n\rangle\langle n|\partial_{k_i} H|n\rangle}{(\epsilon_n - \epsilon_m)^2} \\
& + \sum_{g}{}' f(\epsilon_n) \frac{\langle n|\partial_{k_j} H|n\rangle\langle n|\partial_{k_r} H|g\rangle\langle g|\partial_{k_i} H|n\rangle}{(\epsilon_n - \epsilon_g)^2} + \frac{1}{2} e^2 f(\epsilon_n) \langle n|\partial_{k_r}\partial_{k_i}\partial_{k_j} H|n\rangle \Bigg].
\end{aligned} \tag{4.76}$$

Here $\sum_g'$ ($\sum_{m,g}'$) denotes the summation where the band index g (m, g) runs over those that do not set the denominator to zero. We note that the 5th to 8th terms are obtained by setting one energy denominator to be $1/\omega$ and expanding the other energy denominator up to ω^2 in the 2nd and the 4th terms in Eq. (4.75). In addition, the time reversal symmetry, $\mathcal{T} = K$, leads to the symmetry properties of the Hamiltonian and its eigenstates given by

$$H(k) = H(-k), \qquad \epsilon(k) = \epsilon(-k), \qquad |n(k)\rangle = \langle n(-k)|. \tag{4.77}$$

By using these properties that hold in the presence of the time-reversal symmetry, we find that the above expression for J_r^0 vanishes in the zeroth order. Therefore, the lowest order term is actually the first order term J_r^1.

The first order term in ω is written as

$$J_r = -e^3\omega \sum_{i,j} A_i A_j \int [d\mathbf{k}] \sum_n \Bigg[\sum_g{}' (f(\epsilon_n) - f(\epsilon_g)) \frac{\langle n|\partial_{k_r} H|g\rangle\langle g|\partial_{k_i}\partial_{k_j} H|n\rangle}{(\epsilon_n - \epsilon_g)^2}$$

$$
\begin{aligned}
&+2\sum_{m,g}{}' \left(\frac{f(\epsilon_n)}{\epsilon_n-\epsilon_m}-\frac{f(\epsilon_g)}{\epsilon_g-\epsilon_m}\right)\frac{\langle n|\partial_{k_r}H|g\rangle\langle g|\partial_{k_i}H|m\rangle\langle m|\partial_{k_j}H|n\rangle}{(\epsilon_n-\epsilon_g)^2}\\
&+\sum_{m,g}{}' \left(\frac{f(\epsilon_n)}{(\epsilon_n-\epsilon_m)^2}+\frac{f(\epsilon_g)}{(\epsilon_g-\epsilon_m)^2}\right)\frac{\langle n|\partial_{k_r}H|g\rangle\langle g|\partial_{k_i}H|m\rangle\langle m|\partial_{k_j}H|n\rangle}{\epsilon_n-\epsilon_g}\\
&-4\sum_{g}{}' f(\epsilon_n)\frac{\langle n|\partial_{k_r}H|g\rangle\langle g|\partial_{k_i}H|n\rangle\langle n|\partial_{k_j}H|n\rangle}{(\epsilon_n-\epsilon_g)^3}\\
&-4\sum_{g}{}' f(\epsilon_g)\frac{\langle n|\partial_{k_r}H|g\rangle\langle g|\partial_{k_i}H|g\rangle\langle g|\partial_{k_j}H|n\rangle}{(\epsilon_n-\epsilon_g)^3}\\
&+\sum_{g}{}' (f(\epsilon_n)-f(\epsilon_g))\frac{\langle n|\partial_{k_r}\partial_{k_j}H|g\rangle\langle g|\partial_{k_i}H|n\rangle}{(\epsilon_n-\epsilon_g)^2}\\
&-\sum_{m}{}' f(\epsilon_n)\frac{\langle n|\partial_{k_j}H|m\rangle\langle m|\partial_{k_r}H|n\rangle\langle n|\partial_{k_i}H|n\rangle}{(\epsilon_n-\epsilon_m)^3}\\
&+\sum_{g}{}' f(\epsilon_n)\frac{\langle n|\partial_{k_j}H|n\rangle\langle n|\partial_{k_r}H|g\rangle\langle g|\partial_{k_i}H|n\rangle}{(\epsilon_n-\epsilon_g)^3}\\
&-\sum_{m,g}{}' f(\epsilon_n)\frac{\langle n|\partial_{k_j}H|m\rangle\langle m|\partial_{k_r}H|g\rangle\langle g|\partial_{k_i}H|n\rangle}{(\epsilon_n-\epsilon_g)(\epsilon_n-\epsilon_m)^2}\\
&+\sum_{m,g}{}' f(\epsilon_n)\frac{\langle n|\partial_{k_j}H|m\rangle\langle m|\partial_{k_r}H|g\rangle\langle g|\partial_{k_i}H|n\rangle}{(\epsilon_n-\epsilon_g)^2(\epsilon_n-\epsilon_m)}\Bigg].
\end{aligned}
\tag{4.78}
$$

By using the properties from the time reversal symmetry, this can be reduced as

$$
\begin{aligned}
J_r=-2e^3\omega\sum_{i,j}A_iA_j\int[d\mathbf{k}]\sum_n f(\epsilon_n)\Bigg[&\sum_g{}' f(\epsilon_n)\frac{\langle n|\partial_{k_r}H|g\rangle\langle g|\partial_{k_i}\partial_{k_j}H|n\rangle}{(\epsilon_n-\epsilon_g)^2}\\
&+2\sum_{m,g}{}'\frac{1}{\epsilon_n-\epsilon_m}\frac{\langle n|\partial_{k_r}H|g\rangle\langle g|\partial_{k_i}H|m\rangle\langle m|\partial_{k_j}H|n\rangle}{(\epsilon_n-\epsilon_g)^2}\\
&+\sum_{m,g}{}'\frac{1}{(\epsilon_n-\epsilon_m)^2}\frac{\langle n|\partial_{k_r}H|g\rangle\langle g|\partial_{k_i}H|m\rangle\langle m|\partial_{k_j}H|n\rangle}{\epsilon_n-\epsilon_g}\\
&-3\sum_{g}{}'\frac{\langle n|\partial_{k_r}H|g\rangle\langle g|\partial_{k_i}H|n\rangle\langle n|\partial_{k_j}H|n\rangle}{(\epsilon_n-\epsilon_g)^3}\\
&+\sum_{g}{}'\frac{\langle n|\partial_{k_r}\partial_{k_j}H|g\rangle\langle g|\partial_{k_i}H|n\rangle}{(\epsilon_n-\epsilon_g)^2}-\sum_{m,g}{}'\frac{\langle n|\partial_{k_j}H|m\rangle\langle m|\partial_{k_r}H|g\rangle\langle g|\partial_{k_i}H|n\rangle}{(\epsilon_n-\epsilon_g)(\epsilon_n-\epsilon_m)^2}\Bigg].
\end{aligned}
\tag{4.79}
$$

Now let us consider the specific case relevant to the Berry curvature dipole formula. Namely we suppose that E is applied along the x-direction and consider the current J in the y-direction:

$$\begin{aligned}
J_y = -2e^3\omega A_x A_x \int [d\mathbf{k}] \sum_n f(\epsilon_n) \Bigg[\sum_g{}' f(\epsilon_n) \frac{\langle n|\partial_{k_y} H|g\rangle\langle g|\partial_{k_x}\partial_{k_x} H|n\rangle}{(\epsilon_n-\epsilon_g)^2} \\
+2\sum_{m,g}{}' \frac{1}{\epsilon_n-\epsilon_m} \frac{\langle n|\partial_{k_y} H|g\rangle\langle g|\partial_{k_x} H|m\rangle\langle m|\partial_{k_x} H|n\rangle}{(\epsilon_n-\epsilon_g)^2} \\
-3\sum_g{}' \frac{\langle n|\partial_{k_y} H|g\rangle\langle g|\partial_{k_x} H|n\rangle\langle n|\partial_{k_x} H|n\rangle}{(\epsilon_n-\epsilon_g)^3} \\
+\sum_{m,g}{}' \frac{1}{(\epsilon_n-\epsilon_m)^2} \frac{\langle n|\partial_{k_y} H|g\rangle\langle g|\partial_{k_x} H|m\rangle\langle m|\partial_{k_x} H|n\rangle}{\epsilon_n-\epsilon_g} \\
+\sum_g{}' \frac{\langle n|\partial_{k_y}\partial_{k_x} H|g\rangle\langle g|\partial_{k_x} H|n\rangle}{(\epsilon_n-\epsilon_g)^2} - \sum_{m,g}{}' \frac{\langle n|\partial_{k_x} H|m\rangle\langle m|\partial_{k_y} H|g\rangle\langle g|\partial_{k_x} H|n\rangle}{(\epsilon_n-\epsilon_g)(\epsilon_n-\epsilon_m)^2} \Bigg]. \quad (4.80)
\end{aligned}$$

The $\boldsymbol{k}$-integral of the Berry curvature dipole $\Omega_{z,n}$ for the nth band is explicitly written in many band systems as

$$\begin{aligned}
\int [d\mathbf{k}]\ \partial_x \Omega_{z,n}(k) &= -2\partial_x \int [d\mathbf{k}]\ \mathrm{Im}[\langle \partial_x n|\partial_y n\rangle] \\
&= i\partial_x \int [d\mathbf{k}]\ \sum_g [\langle \partial_x n|g\rangle\langle g|\partial_y n\rangle - \langle \partial_y n|g\rangle\langle g|\partial_x n\rangle] \\
&= i\partial_x \int [d\mathbf{k}]\ \sum_g{}' \left[\frac{\langle n|\partial_x H|g\rangle\langle g|\partial_y H|n\rangle}{(\epsilon_n-\epsilon_g)^2} - \frac{\langle n|\partial_y H|g\rangle\langle g|\partial_x H|n\rangle}{(\epsilon_n-\epsilon_g)^2} \right] \\
&= -2i \int [d\mathbf{k}]\ \sum_g{}' \Bigg[\frac{\langle n|\partial_y H|g\rangle\langle \partial_x g|\partial_x H|n\rangle}{(\epsilon_n-\epsilon_g)^2} + \frac{\langle n|\partial_y H|g\rangle\langle g|\partial_x\partial_x H|n\rangle}{(\epsilon_n-\epsilon_g)^2} \\
&\quad + \frac{\langle n|\partial_y H|g\rangle\langle g|\partial_x H|\partial_x n\rangle}{(\epsilon_n-\epsilon_g)^2} + \frac{\langle \partial_x n|\partial_y H|g\rangle\langle g|\partial_x H|n\rangle}{(\epsilon_n-\epsilon_g)^2} \\
&\quad + \frac{\langle n|\partial_x\partial_y H|g\rangle\langle g|\partial_x H|n\rangle}{(\epsilon_n-\epsilon_g)^2} + \frac{\langle n|\partial_y H|\partial_x g\rangle\langle g|\partial_x H|n\rangle}{(\epsilon_n-\epsilon_g)^2} \\
&\quad - 2\frac{\langle n|\partial_y H|g\rangle\langle g|\partial_x H|n\rangle}{(\epsilon_n-\epsilon_g)^3}[(v_x)_{nn}-(v_x)_{gg}] \Bigg] \\
&= -2i \int [d\mathbf{k}]\ \sum_g{}' \Bigg[\frac{\langle n|\partial_y H|g\rangle\langle g|\partial_x\partial_x H|n\rangle}{(\epsilon_n-\epsilon_g)^2} + \frac{\langle n|\partial_x\partial_y H|g\rangle\langle g|\partial_x H|n\rangle}{(\epsilon_n-\epsilon_g)^2} \\
&\quad - 2\frac{\langle n|\partial_y H|g\rangle\langle g|\partial_x H|n\rangle}{(\epsilon_n-\epsilon_g)^3}[(v_x)_{nn}-(v_x)_{gg}] \Bigg]
\end{aligned}$$

$$+\sum_{g,m}{}' \Bigg[-\frac{\langle n|\partial_y H|g\rangle\langle g|\partial_x H|m\rangle\langle m|\partial_x H|n\rangle}{(\epsilon_m-\epsilon_g)(\epsilon_n-\epsilon_g)^2} + \frac{\langle n|\partial_y H|g\rangle\langle g|\partial_x H|m\rangle\langle m|\partial_x H|n\rangle}{(\epsilon_n-\epsilon_m)(\epsilon_n-\epsilon_g)^2} - \frac{\langle n|\partial_x H|m\rangle\langle m|\partial_y H|g\rangle\langle g|\partial_x H|n\rangle}{(\epsilon_m-\epsilon_n)(\epsilon_n-\epsilon_g)^2} + \frac{\langle n|\partial_y H|g\rangle\langle g|\partial_x H|m\rangle\langle m|\partial_x H|n\rangle}{(\epsilon_m-\epsilon_g)(\epsilon_n-\epsilon_m)^2} \Bigg], \tag{4.81}$$

where we have used the time reversal symmetry to simplify the expressions and the equation $\langle n|\partial_k m\rangle = \langle n|v|m\rangle/(\epsilon_m-\epsilon_n)$. We note that the region of the above $\boldsymbol{k}$-integration can be any $\mathcal{T}$-symmetric region that includes both $\boldsymbol{k}$ and $-\boldsymbol{k}$, especially, the Fermi sea satisfying $f(\epsilon_n)=1$.

By using Eq.(4.81), we finally obtain

$$\begin{aligned} J_y = -2e^3\omega A_x A_x \int [d\mathbf{k}] \sum_n f(\epsilon_n) \Bigg[& \frac{\partial_x \Omega_{z,n}}{-2i} + \sum_g{}' \frac{\langle n|\partial_y H|g\rangle\langle g|\partial_x H|n\rangle\langle n|\partial_x H|n\rangle}{(\epsilon_n-\epsilon_g)^3} \\ & + \sum_g{}' \frac{1}{(\epsilon_n-\epsilon_g)^2} \frac{\langle n|\partial_{k_y} H|g\rangle\langle g|\partial_{k_x} H|g\rangle\langle g|\partial_{k_x} H|n\rangle}{\epsilon_n-\epsilon_g} \\ & + \sum_g{}' \frac{1}{\epsilon_n-\epsilon_g} \frac{\langle n|\partial_{k_y} H|g\rangle\langle g|\partial_{k_x} H|g\rangle\langle g|\partial_{k_x} H|n\rangle}{(\epsilon_n-\epsilon_g)^2} \\ & - 3\sum_g{}' \frac{\langle n|\partial_{k_y} H|g\rangle\langle g|\partial_{k_x} H|n\rangle\langle n|\partial_{k_x} H|n\rangle}{(\epsilon_n-\epsilon_g)^3} \\ & + 2\sum_g{}' \frac{\langle n|\partial_y H|g\rangle\langle g|\partial_x H|n\rangle}{(\epsilon_n-\epsilon_g)^3} [(v_x)_{nn} - (v_x)_{gg}] \Bigg] \\ = -ie^3\omega A_x A_x \int [d\mathbf{k}] \ & \partial_x \Omega_z. \end{aligned} \tag{4.82}$$

This indicates that the nonlinear conductivity for the SHG is given by

$$\sigma_{yxx} = \frac{ie^3}{\omega} \int [d\mathbf{k}] \sum_n f(\epsilon_n) \partial_{k_x} \Omega_{z,n}, \tag{4.83}$$

which reproduces Eq. (4.44) in Sec. 4.2B and proves the Berry curvature dipole formula for SHG in general cases with many bands.

Chapter 5

Hall conductivity in an inhomogeneous electric field

In this chapter we will study the Hall conductivity of insulators in a static inhomogeneous electric field to the second order of wave number q using the Kubo formula. We derive general formulas for the q^2 term in hall conductivity. We find that for an insulator with only two bands that are flat, the q^2 term is determined by the quantum metric tensor and the Berry curvature. Using a semiclassical derivation we provide an intuitive picture of how the quantum metric tensor appears. We end this chapter with numerical studies on a topological flat band model and the Hofstadter model.

5.1 Preliminaries and background

Strain tensor [45]Under deformation the position x of a point in a solid body will be mapped to a new position x'. We use Euclidean coordinates $\{x_i\}$ to label the original position of the point and the deformation will be equivalent to a change of the metric of the coordinate system. In the new Euclidean coordinate system the distance between two very close points will be

$$dl'^2 = \sum_i dx_i'^2 = \sum_i (\frac{\partial x_i'}{\partial x_j} dx_j)^2 = \sum_i \frac{\partial x_i'}{\partial x_j}\frac{\partial x_i'}{\partial x_k} dx_j dx_k \,. \tag{5.1}$$

We can rewrite it in terms of the displacement of the point $u_i = x_i' - x_i$,

$$dl'^2 = dl^2 + 2u_{ik} dx_i dx_k \tag{5.2}$$

where $u_{ik} = \frac{1}{2}\,(\frac{\partial u_i}{\partial x_k} + \frac{\partial u_k}{\partial x_i} + \frac{\partial u_l}{\partial x_i}\frac{\partial u_l}{\partial x_k}\,)$ is the strain tensor. Usually the deformation we consider is very small so we can neglect the last term in the strain tensor u_{ik} and have

$$u_{ik} = \frac{1}{2}\left(\frac{\partial u_i}{\partial x_k} + \frac{\partial u_k}{\partial x_i}\right) \tag{5.3}$$

Stress tensor When deformation occurs forces will arise as they would like to make the system return to equilibrium. These forces are called internal stresses and in our assumption they have very short range of action.

Let's consider one component of the total force caused by deformation on some portion of the system. It will be given by

$$\int F_i(\vec{x})dV \tag{5.4}$$

where the integral is taken over the volume of that portion of the body. Since the stress is short range and hence can only act on the surface of the body, the integral should be able to be rewritten as an integral over a surface. Therefore, there must exist a tensor σ_{ik} such that F_i is given by

$$F_i = \frac{\partial \sigma_{ik}}{\partial x_k}\,. \tag{5.5}$$

The tensor σ_{ik} here is called the stress tensor.

Viscosity The deformation of a body will be reversible if it occurs with an infinitesimal speed so that the thermodynamic equilibrium is established at every moment. In the process of realistic deformation if there is any internal motion in the body, there will be irreversible processes arising from the finite velocity of that motion. The irreversible processes will then lead to energy dissipation. This means of dissipation is called viscosity.

It is clear that the forces in the irreversible process should relate to the velocity of the deformation. In addition, it should meet the requirement that when the motion of the system is a general translation or rotation the forces should be zero as there is no internal motion. It turns out that the dissipation can be described by the time derivative of the strain tensor $\dot{\mu}$.

Including the forces from viscosity the stress tensor will be given by

$$\sigma_{\alpha\beta} = \lambda_{\alpha\beta\gamma\delta}\mu_{\gamma\delta} - \eta_{\alpha\beta\gamma\delta}\dot{\mu}_{\gamma\delta} \tag{5.6}$$

where the first coefficient is called elastic modulus and the second is the viscosity tensor.

Odd viscosity In general, the viscosity tensor need not to be symmetric. Actually one the symmetric part of the viscosity tensor describes the dissipation while the antisymmetric part is not associated with dissipation. We call the antisymmetric part odd viscosity and it vanishes if there is time reversal symmetry according to Onsager relation.

$$\begin{aligned} \eta^S(B) =& \eta^S(-B) \\ \eta^A(B) =& -\eta^A(-B) \end{aligned} \tag{5.7}$$

In two dimensions, η^A has three independent coefficients $\eta^A_{1112}, \eta^A_{1122}, \eta^A_{1222}$. In an isotropic system we have

$$\eta^A_{1112} = \eta^A_{1222} = \eta, \quad \eta^A_{1122} = 0\,. \tag{5.8}$$

Hall viscosity Quantum fluids have a ground state with a finite gap. At zero temperature they have nondissipative responses so we will have $\eta^S = 0$ while η^A may or may not be zero.

We now consider a family of Hamiltonians $H(u_{\alpha\beta})$ which depends smoothly on $u_{\alpha\beta}$. According to the adiabatic response theory, in a uniform deformation we will have [8]

$$\left\langle \frac{\partial H}{\partial u_{\alpha\beta}} \right\rangle = \frac{\partial \epsilon}{\partial u_{\alpha\beta}} + \sum_{\lambda\delta} \Omega_{\alpha\beta\lambda\delta} \dot{u}_{\lambda\delta} \,. \tag{5.9}$$

where ϵ is the energy of the system. The adiabatic curvature Ω is the corresponding non-dissipative (odd) viscosity tensor and it is also named as Hall viscosity.

Hall conductivity and Hall viscosity In a Galilean invariant system in which particles have the same charge/mass ratio, it has been shown that Hall viscosity is related to the Hall conductivity in an inhomogeneous electric field [37, 15]. It is shown that in a two dimensional quantum Hall system, the Hall conductivity tensor $\sigma_{xy}(q)$ can be expanded as

$$\frac{\sigma_{xy}(q)}{\sigma_{xy}(0)} = 1 + C_2(q^2\ell^2) + \mathcal{O}(q^4\ell^4) \tag{5.10}$$

where $\ell = \sqrt{\hbar c/eB}$ is the magnetic length and

$$C_2 = \frac{\eta^A}{\hbar n} - \frac{2\pi}{\nu}\frac{\ell^2}{\hbar\omega_c} B^2 \epsilon''(B) \tag{5.11}$$

where ν is the filling factor, ϵ is the energy density, n is the density of electrons and $\omega_c = \frac{eB}{mc}$ is the cyclotron frequency of the quantum Hall state. For integer quantum Hall state with $\nu = 1$ the first term gives $\frac{1}{4}$ and the second term gives -1 and in total we will have $C_2 = -\frac{3}{4}$ [37]. In the following sections we will calculate the coefficient of the Hall conductivity in the second order in q using Kubo formula and semiclassical theory.

5.2 Derivation of the q^2 term in Hall conductivity

5.2.1 Symmetry analysis

Before we do the derivation we would like to first do the symmetry analysis for the Hall conductivity tensor. We do it by looking at requirements on the dielectric tensor $\epsilon_{ij}(\omega, \mathbf{q})$ and using the relation between the dielectric tensor and the Hall conductivity tensor, $\sigma_{ij}(\omega, \mathbf{q}) = i\omega\epsilon_{ij}(\omega, \mathbf{q})$. The dielectric tensor for a non-dissipative system obeys the following three requirements:

- Hermitian: $\epsilon_{ij}(\omega, \mathbf{q}) = \epsilon^*_{ji}(\omega, \mathbf{q})$
- Onsager's relation: $\epsilon_{ij}(\omega, \mathbf{q}, B) = \epsilon_{ji}(\omega, -\mathbf{q}, -B)$

- Eq.(103.9) in Landau's book of Electrodynamics [44]: $\epsilon_{ij}(\omega,\mathbf{q}) = \epsilon^*_{ij}(-\omega,-\mathbf{q})$.

By expanding those equations to the second order in q $\epsilon_{ij}(\omega,\mathbf{q}) = \epsilon^0_{ij}(\omega) + \epsilon^1_{ijl}q_l(\omega) + \epsilon^2_{ijlg}q_lq_g(\omega)$ and comparing coefficients at the same order of q we get:

- Hermitian: $\epsilon^1_{ijl}(\omega) = \epsilon^{1*}_{jil}(\omega)$, $\epsilon^2_{ijlg}(\omega) = \epsilon^{2*}_{jilg}(\omega)$
- Onsager's relation: $\epsilon^1_{ijl}(\omega,B) = -\epsilon^1_{jil}(\omega,-B)$, $\epsilon^2_{ijlg}(\omega,B) = \epsilon^2_{jilg}(\omega,-B)$
- Eq.(103.9) in Landau's book of Electrodynamics: $\epsilon^1_{ijl}(\omega) = -\epsilon^{1*}_{ijl}(-\omega)$, $\epsilon^2_{ijlg}(\omega) = \epsilon^{2*}_{ijlg}(-\omega)$

The dielectric tensor can be split into two parts: time reversal even $\epsilon^S_{ij}(B) = \epsilon^S_{ij}(-B)$ and time reversal odd $\epsilon^A_{ij}(B) = -\epsilon^A_{ij}(-B)$. Since we would like to work on the Hall conductivity we look at the time reversal odd part. The above equations will give

$$\begin{array}{ccccccc} \epsilon^{A1}_{ijl}(\omega) & = & -\epsilon^{A1*}_{ijl}(-\omega) & = & \epsilon^{A1}_{jil}(\omega) & = & \epsilon^{A1*}_{jil}(\omega) \\ \epsilon^{2A}_{ijlg}(\omega) & = & \epsilon^{A2*}_{ijlg}(-\omega) & = & -\epsilon^{A2}_{jilg}(\omega) & = & \epsilon^{A2*}_{jilg}(\omega) \end{array} \tag{5.12}$$

It indicates that ϵ^{A1}_{ijl} is real, odd in ω, and symmetric in $i \leftrightarrow j$ and that ϵ^{A2}_{ijlg} is imaginary, odd in ω, and antisymmetric in $i \leftrightarrow j$. Similarly one can see that ϵ^{1S}_{ijl} is imaginary, even in ω, and antisymmetric in $i \leftrightarrow j$ and ϵ^{S2}_{ijlg} is real, even in ω, and symmetric in $i \leftrightarrow j$. Note that the Hall conductivity tensor $\sigma_{ij}(\mathbf{q})$ for a static inhomogeneous electric field would be even in ω and antisymmetric in $i \leftrightarrow j$. Therefore, we reach the conclusion that the next higher order correction to the zeroth order hall conductivity σ^0_{ij} is

$$\sigma^{A2}_{ijlg}q_lq_g = \lim_{\omega\to 0} i\omega\epsilon^{A2}_{ijlg}(\omega)q_lq_g\,. \tag{5.13}$$

5.2.2 Derivation using Kubo formula

In this section, we use Kubo formula to calculate the Hall conductivity $\sigma_{ij}(\mathbf{q})$ for an insulator. The Hamiltonian we consider includes only one particle potential. The current response to a vector potential $\mathbf{A}(\omega,\mathbf{q})$ is given by

$$\begin{aligned} J_i(\omega,\mathbf{q}) = &-e^2\int[d\mathbf{k}]\sum_{n,m}\frac{f(\epsilon_{n,\mathbf{k}-\mathbf{q}/2}) - f(\epsilon_{m,\mathbf{k}+\mathbf{q}/2})}{\epsilon_{n,\mathbf{k}-\mathbf{q}/2} - \epsilon_{m,\mathbf{k}+\mathbf{q}/2} + \omega} \\ &\langle n_{\mathbf{k}-\mathbf{q}/2}|\partial_i H|m_{\mathbf{k}+\mathbf{q}/2}\rangle\langle m_{\mathbf{k}+\mathbf{q}/2}|\partial_j H|n_{\mathbf{k}-\mathbf{q}/2}\rangle A_j(\omega,\mathbf{q}) \end{aligned} \tag{5.14}$$

where ϵ_n is the band energy, $|n\rangle$ is short for $|u_{nk}\rangle$ and $A_j(\omega,\mathbf{q}) = A_j e^{-iwt+i\mathbf{q}\cdot\mathbf{r}}$. Since we work on insulators, the distribution function $f(\epsilon_n,\mathbf{k}+\mathbf{q}/2) = f(\epsilon_n)$ is 1 for occupied bands and 0 for unoccupied bands. As we work on Hall conductivity tensor we focus on the antisymmetric part of the conductivity tensor and have

$$\begin{aligned} J_i(\omega) = &-e^2\int[d\mathbf{k}]\sum_{n,m}\frac{f(\epsilon_n) - f(\epsilon_m)}{\epsilon_{n,\mathbf{k}-\mathbf{q}/2} - \epsilon_{m,\mathbf{k}+\mathbf{q}/2} + \omega} \\ &\frac{1}{2}\,\mathrm{Im}\left[\langle n_{\mathbf{k}-\mathbf{q}/2}|\partial_i H|m_{\mathbf{k}+\mathbf{q}/2}\rangle\langle m_{\mathbf{k}+\mathbf{q}/2}|\partial_j H|n_{\mathbf{k}-\mathbf{q}/2}\rangle - (i\leftrightarrow j)\right]A_j(\omega,\mathbf{q})\,. \end{aligned} \tag{5.15}$$

In order to get the q^2 term we would like to expand it in terms of $\mathbf{q}$ and ω. For simplicity we assume $\mathbf{q}$ has only one component q_l

$$\begin{aligned}\frac{1}{\epsilon_{n,\mathbf{k}-\mathbf{q}/2}-\epsilon_{m,\mathbf{k}+\mathbf{q}/2}+\omega} =& \frac{1}{\epsilon_{n,\mathbf{k}-\mathbf{q}/2}-\epsilon_{m,\mathbf{k}+\mathbf{q}/2}}-\frac{\omega}{(\epsilon_n-\epsilon_m)^2}-\frac{\omega q_l\left(V_{nl}+V_{ml}\right)}{(\epsilon_n-\epsilon_m)^3}\\ &-\frac{\omega q_l^2 3\left(V_{nl}+V_{ml}\right)^2}{4(\epsilon_n-\epsilon_m)^4}+\frac{\omega q_l^2\left(\partial_l V_{nl}-\partial_l V_{ml}\right)}{4(\epsilon_n-\epsilon_m)^3}+\cdots\end{aligned} \tag{5.16}$$

where $V_{nl}=\partial_{k_l}\epsilon_n(\mathbf{k})$ is the band velocity. We have omitted the $\mathbf{k}$ dependence in the above quantities. And the other part is also expanded:

$$\begin{aligned}&\langle n_{\mathbf{k}-\mathbf{q}/2}|\partial_i H|m_{\mathbf{k}+\mathbf{q}/2}\rangle\langle m_{\mathbf{k}+\mathbf{q}/2}|\partial_j H|n_{\mathbf{k}-\mathbf{q}/2}\rangle = \langle n|\partial_i H|m\rangle\langle m|\partial_j H|n\rangle\\ &+\frac{q_l}{2}\Big(\quad i\,\mathrm{Im}[\langle n|\partial_i H|\partial_l m\rangle\langle m|\partial_j H|n\rangle-\langle\partial_l n|\partial_i H|m\rangle\langle m|\partial_j H|n\rangle-(i\leftrightarrow j)]\\ &\qquad+\mathrm{Re}[\langle n|\partial_i H|\partial_l m\rangle\langle m|\partial_j H|n\rangle-\langle\partial_l n|\partial_i H|m\rangle\langle m|\partial_j H|n\rangle+(i\leftrightarrow j)]\Big)\\ &+\frac{q_l q_l}{4}\Big(\quad i\,\mathrm{Im}[\,\frac{1}{2}\langle n|\partial_i H|\partial_l m\rangle\langle\partial_l m|\partial_j H|n\rangle-\langle\partial_l n|\partial_i H|m\rangle\langle\partial_l m|\partial_j H|n\rangle\\ &\qquad\qquad-\langle\partial_l n|\partial_i H|\partial_l m\rangle\langle m|\partial_j H|n\rangle+\frac{1}{2}\langle\partial_l n|\partial_i H|m\rangle\langle m|\partial_j H|\partial_l n\rangle-(i\leftrightarrow j)]\\ &\qquad+\frac{1}{2}i\,\mathrm{Im}[\langle n|\partial_i H|\partial_l^2 m\rangle\langle m|\partial_j H|n\rangle+\langle\partial_l^2 n|\partial_i H|m\rangle\langle m|\partial_j H|n\rangle-(i\leftrightarrow j)]\\ &\qquad+\mathrm{Re}[\,\frac{1}{2}\langle n|\partial_i H|\partial_l m\rangle\langle\partial_l m|\partial_j H|n\rangle-\langle\partial_l n|\partial_i H|m\rangle\langle\partial_l m|\partial_j H|n\rangle\\ &\qquad\qquad-\langle\partial_l n|\partial_i H|\partial_l m\rangle\langle m|\partial_j H|n\rangle+\frac{1}{2}\langle\partial_l n|\partial_i H|m\rangle\langle m|\partial_j H|\partial_l n\rangle+(i\leftrightarrow j)]\\ &\qquad+\frac{1}{2}\,\mathrm{Re}[\langle n|\partial_i H|\partial_l^2 m\rangle\langle m|\partial_j H|n\rangle+\langle\partial_l^2 n|\partial_i H|m\rangle\langle m|\partial_j H|n\rangle+(i\leftrightarrow j)]\Big)\end{aligned} \tag{5.17}$$

The current which is first order in ω and second order in $\mathbf{q}$ is given by

$$\begin{aligned}J_i^{(2)} =& -e^2(\partial_{x_l}\partial_{x_l}E_j)\int[d\mathbf{k}]\sum_{n,m\neq n}f(\epsilon_n)\Big\{\\ &\left(\frac{\partial_l V_{nl}-\partial_l V_{ml}}{2(\epsilon_m-\epsilon_n)^3}+\frac{3(V_{nl}+V_{ml})^2}{2(\epsilon_m-\epsilon_n)^4}\right)\mathrm{Im}\langle n|\partial_i H|m\rangle\langle m|\partial_j H|n\rangle\\ &-\frac{V_{nl}+V_{ml}}{(\epsilon_m-\epsilon_n)^3}\,\mathrm{Im}[\langle n|\partial_i H|\partial_l m\rangle\langle m|\partial_j H|n\rangle-\langle\partial_l n|\partial_i H|m\rangle\langle m|\partial_j H|n\rangle-(i\leftrightarrow j)]\\ &+\frac{1}{2(\epsilon_m-\epsilon_n)^2}\Big(\quad\mathrm{Im}[\frac{1}{2}\langle n|\partial_i H|\partial_l m\rangle\langle\partial_l m|\partial_j H|n\rangle-\langle\partial_l n|\partial_i H|m\rangle\langle\partial_l m|\partial_j H|n\rangle\\ &\qquad\qquad-\langle\partial_l n|\partial_i H|\partial_l m\rangle\langle m|\partial_j H|n\rangle+\frac{1}{2}\langle\partial_l n|\partial_i H|m\rangle\langle m|\partial_j H|\partial_l n\rangle-(i\leftrightarrow j)]\\ &\qquad+\frac{1}{2}\,\mathrm{Im}[\langle n|\partial_i H|\partial_l^2 m\rangle\langle m|\partial_j H|n\rangle+\langle\partial_l^2 n|\partial_i H|m\rangle\langle m|\partial_j H|n\rangle-(i\leftrightarrow j)]\Big)\Big\},\end{aligned} \tag{5.18}$$

To simplify the derivation we first work on a two band model with v and c denote the valence band and the conduction band respectively. The current for a two band model is given by

$$\begin{aligned}J_i^{(2)} = -e^2(\partial_{x_l}\partial_{x_l}E_j)\int[d\mathbf{k}]\Big\{ &\left(\frac{\partial_l V_{vl}-\partial_l V_{cl}}{2(\epsilon_c-\epsilon_v)^3}+\frac{3(V_{vl}+V_{cl})^2}{2(\epsilon_c-\epsilon_v)^4}\right)\mathrm{Im}\langle v|\partial_i H|c\rangle\langle c|\partial_j H|v\rangle\\ &-\frac{V_{vl}+V_{cl}}{(\epsilon_c-\epsilon_v)^3}\,\mathrm{Im}[\langle v|\partial_i H|\partial_l c\rangle\langle c|\partial_j H|v\rangle-\langle\partial_l v|\partial_i H|c\rangle\langle c|\partial_j H|v\rangle-(i\leftrightarrow j)]\\ &+\frac{1}{2(\epsilon_c-\epsilon_v)^2}\Big(\mathrm{Im}[\frac{1}{2}\langle v|\partial_i H|\partial_l c\rangle\langle\partial_l c|\partial_j|v\rangle-\langle\partial_l v|\partial_i H|c\rangle\langle\partial_l c|\partial_j H|v\rangle\\ &-\langle\partial_l v|\partial_i H|\partial_l c\rangle\langle c|\partial_j H|v\rangle+\frac{1}{2}\langle\partial_l v|\partial_i H|c\rangle\langle c|\partial_j H|\partial_l v\rangle-(i\leftrightarrow j)]\\ &+\frac{1}{2}\,\mathrm{Im}\left[\langle v|\partial_i H|\partial_l^2 c\rangle\langle c|\partial_j H|v\rangle+\langle\partial_l^2 v|\partial_i H|c\rangle\langle c|\partial_j H|v\rangle-(i\leftrightarrow j)\right]\Big)\Big\}\,.\end{aligned} \tag{5.19}$$

We then use

$$\langle n|\partial_i H|m\rangle = \delta^n_m V_{ni}+(\epsilon_m-\epsilon_n)\langle n|\partial_i m\rangle \tag{5.20}$$

and insert $|v\rangle\langle v|+|c\rangle\langle c|$ in proper positions in the first line in Eq.(5.19) to get

$$\mathrm{Im}\langle v|\partial_i H|c\rangle\langle c|\partial_j H|v\rangle = (\epsilon_c-\epsilon_v)^2\,\mathrm{Im}\langle\partial_i v|\partial_j v\rangle\,. \tag{5.21}$$

We do the same thing for the second line in Eq.(5.19)

$$\begin{aligned}&\mathrm{Im}[\langle v|\partial_i H|\partial_l c\rangle\langle c|\partial_j H|v\rangle-\langle\partial_l v|\partial_i H|c\rangle\langle c|\partial_j H|v\rangle-(i\leftrightarrow j)]\\ &=(\epsilon_v-\epsilon_c)\{\mathrm{Im}\,\partial_l[\langle v|\partial_i c\rangle\langle c|\partial_j v\rangle(\epsilon_v-\epsilon_c)]+\partial_j[\langle v|\partial_l c\rangle\langle c|\partial_i v\rangle(\epsilon_v-\epsilon_c)]\\ &\quad-\partial_i[\langle v|\partial_l c\rangle\langle c|\partial_j v\rangle(\epsilon_v-\epsilon_c)]+\mathrm{Im}[(V_{vl}+V_{cl})\langle v|\partial_i c\rangle\langle c|\partial_j v\rangle]\\ &\quad+2\,\mathrm{Im}[V_{vl}\langle\partial_i v|c\rangle\langle c|\partial_j v\rangle+V_{vj}\langle\partial_l v|c\langle c|\partial_i v\rangle-V_{vi}\langle\partial_l v|c\rangle\langle c|\partial_j v\rangle]\}\\ &=-(\epsilon_v-\epsilon_c)[(V_{vl}+V_{cl})\,\mathrm{Im}\langle\partial_i v|\partial_j v\rangle+\varepsilon^{ijl}(\vec{V}_v\cdot\Omega_v)-\varepsilon^{ijl}\frac{1}{e}(\nabla\cdot\vec{M}_v)]\,.\end{aligned} \tag{5.22}$$

where we have used the expression for orbital magnetic moment $\vec{M}_v=\frac{e}{2}\,\mathrm{Im}\langle\nabla_{\mathbf{k}}v|\times(H-\epsilon_v)|\nabla_{\mathbf{k}}v\rangle$ and the expression for Berry curvature $\Omega_{vl}=-\varepsilon^{ijl}\,\mathrm{Im}\langle\partial_i v|\partial_j v\rangle$. For the third line in Eq.(5.19) we get

$$\begin{aligned}&\mathrm{Im}[\frac{1}{2}\langle v|\partial_i H|\partial_l c\rangle\langle\partial_l c|\partial_j H|v\rangle-\langle\partial_l v|\partial_i H|c\rangle\langle\partial_l c|\partial_j H|v\rangle\\ &-\langle\partial_l v|\partial_i H|\partial_l c\rangle\langle c|\partial_j H|v\rangle+\frac{1}{2}\langle\partial_l v|\partial_i H|c\rangle\langle c|\partial_j H|\partial_l v\rangle-(i\leftrightarrow j)]\\ &=\mathrm{Im}[(\epsilon_c-\epsilon_v)\langle\partial_l v|c\rangle\langle c|\partial_l c\rangle\langle\partial_j c|v\rangle V_{vi}+(\epsilon_c-\epsilon_v)\langle\partial_l v|v\rangle\langle v|\partial_l c\rangle\langle\partial_j c|v\rangle V_{ci}] &(5.23)\\ &\quad+\frac{1}{2}|\langle\partial_l v|v\rangle|^2\langle\partial_i v|\partial_j v\rangle(\epsilon_c-\epsilon_v)^2+\frac{1}{2}|\langle\partial_l c|c\rangle|^2\langle\partial_i v|\partial_j v\rangle(\epsilon_c-\epsilon_v)^2-(i\leftrightarrow j)] &(5.24)\end{aligned}$$

and

$$\begin{aligned}
&\frac{1}{2}\,\mathrm{Im}\left[\langle v|\partial_i H|\partial_l^2 c\rangle\langle c|\partial_j H|v\rangle + \langle\partial_l^2 v|\partial_i H|c\rangle\langle c|\partial_j H|v\rangle - (i\leftrightarrow j)\right]\\
=&\frac{1}{2}\,\mathrm{Im}[\langle v|\partial_l^2 c\rangle\langle\partial_j c|v\rangle(\epsilon_c-\epsilon_v)V_{vi} + \langle\partial_l^2 v|c\rangle\langle\partial_j c|v\rangle(\epsilon_c-\epsilon_v)V_{ci} && (5.25)\\
&+\langle v|\partial_i c\rangle\langle c|\partial_l^2 c\rangle\langle\partial_j c|v\rangle(\epsilon_c-\epsilon_v)^2 + \langle v|\partial_l^2 v\rangle\langle v|\partial_i c\rangle\langle\partial_j c|v\rangle(\epsilon_c-\epsilon_v)^2 - (i\leftrightarrow j)] && (5.26)\\
=&\frac{1}{2}\,\mathrm{Im}[\langle v|\partial_l^2 c\rangle\langle\partial_j c|v\rangle(\epsilon_c-\epsilon_v)V_{vi} + \langle\partial_l^2 v|c\rangle\langle\partial_j c|v\rangle(\epsilon_c-\epsilon_v)V_{ci} - (i\leftrightarrow j)] && (5.27)\\
&-\langle\partial_l c|\partial_l c\rangle\,\mathrm{Im}[\langle\partial_i v|\partial_j v\rangle](\epsilon_c-\epsilon_v)^2 - \langle\partial_l v|\partial_l v\rangle\,\mathrm{Im}[\langle\partial_i v|\partial_j v\rangle](\epsilon_c-\epsilon_v)^2\,. && (5.28)
\end{aligned}$$

In the last equal sign we have used

$$\begin{aligned}
&\mathrm{Im}[\langle v|\partial_i c\rangle\langle c|\partial_l^2 c\rangle\langle\partial_j c|v\rangle(\epsilon_c-\epsilon_v)^2 - (i\leftrightarrow j)]\\
&\quad=\mathrm{Im}[\langle v|\partial_i c\rangle\langle c|\partial_l^2 c\rangle\langle\partial_j c|v\rangle(\epsilon_c-\epsilon_v)^2 - \langle v|\partial_j c\rangle\langle c|\partial_l^2 c\rangle\langle\partial_i c|v\rangle(\epsilon_c-\epsilon_v)^2]\\
&\quad=(\epsilon_c-\epsilon_v)^2\,\mathrm{Im}[\langle c|\partial_l^2 c\rangle\langle v|\partial_i c\rangle\langle\partial_j c|v\rangle + \langle\partial_l^2 c|c\rangle\langle v|\partial_i c\rangle\langle\partial_j c|v\rangle]\\
&\quad=-2(\epsilon_c-\epsilon_v)^2\,\mathrm{Im}[\langle\partial_l c|\partial_l c\rangle\langle v|\partial_i c\rangle\langle\partial_j c|v\rangle]\\
&\quad=-2(\epsilon_c-\epsilon_v)^2\langle\partial_l c|\partial_l c\rangle\,\mathrm{Im}[\langle\partial_i v|\partial_j v\rangle]\,.
\end{aligned}$$

Similarly,

$$\begin{aligned}
&\mathrm{Im}[\langle v|\partial_l^2 v\rangle\langle v|\partial_i c\rangle\langle\partial_j c|v\rangle(\epsilon_c-\epsilon_v)^2 - (i\leftrightarrow j)]\\
=&-2(\epsilon_c-\epsilon_v)^2\langle\partial_l v|\partial_l v\rangle\,\mathrm{Im}[\langle\partial_i v|\partial_j v\rangle]\,.
\end{aligned} \tag{5.29}$$

Combining Eq.(5.24) and Eq.(5.28) we have

$$\begin{aligned}
&\frac{1}{2}|\langle\partial_l v|v\rangle|^2\,\mathrm{Im}[\langle\partial_i v|\partial_j v\rangle](\epsilon_c-\epsilon_v)^2 + \frac{1}{2}|\langle\partial_l c|c\rangle|^2\,\mathrm{Im}[\langle\partial_i v|\partial_j v\rangle](\epsilon_c-\epsilon_v)^2 - (i\leftrightarrow j)]\\
-&\langle\partial_l c|\partial_l c\rangle\,\mathrm{Im}[\langle\partial_i v|\partial_j v\rangle](\epsilon_c-\epsilon_v)^2 - \langle\partial_l v|\partial_l v\rangle\,\mathrm{Im}[\langle\partial_i v|\partial_j v\rangle](\epsilon_c-\epsilon_v)^2\\
&=(\epsilon_c-\epsilon_v)^2\,\mathrm{Im}[\langle\partial_i v|\partial_j v\rangle]\left(\langle\partial_l v|v\rangle\langle v|\partial_l v\rangle - \langle\partial_l v|v\rangle\langle v|\partial_l v\rangle - \langle\partial_l v|c\rangle\langle c|\partial_l v\rangle + (c\leftrightarrow v)\right)\\
&=(\epsilon_c-\epsilon_v)^2\,\mathrm{Im}[\langle\partial_i v|\partial_j v\rangle]\left(-\langle\partial_l v|c\rangle\langle c|\partial_l v\rangle - \langle\partial_l c|v\rangle\langle v|\partial_l c\rangle\right)\\
&=-2(\epsilon_c-\epsilon_v)^2\,\mathrm{Im}[\langle\partial_i v|\partial_j v\rangle]|\langle\partial_l v|c\rangle|^2.
\end{aligned} \tag{5.30}$$

Combining Eq.(5.23) and Eq.(5.27) we have

$$\begin{aligned}
&\mathrm{Im}[(\epsilon_c-\epsilon_v)\langle\partial_l v|c\rangle\langle c|\partial_l c\rangle\langle\partial_j c|v\rangle V_{vi} + (\epsilon_c-\epsilon_v)\langle\partial_l v|v\rangle\langle v|\partial_l c\rangle\langle\partial_j c|v\rangle V_{ci} - (i\leftrightarrow j)]\\
&+\frac{1}{2}\,\mathrm{Im}[\langle v|\partial_l^2 c\rangle\langle\partial_j c|v\rangle(\epsilon_c-\epsilon_v)V_{vi} + \langle\partial_l^2 v|c\rangle\langle\partial_j c|v\rangle(\epsilon_c-\epsilon_v)V_{ci} - (i\leftrightarrow j)]\\
=&(\epsilon_c-\epsilon_v)\,\mathrm{Im}[\frac{1}{2}\langle\partial_l^2 v|c\rangle\langle\partial_j c|v\rangle(V_{ci}-V_{vi}) + \langle\partial_l v|v\rangle\langle v|\partial_l c\rangle\langle\partial_j c|v\rangle(V_{ci}-V_{vi}) - (i\leftrightarrow j)]
\end{aligned} \tag{5.31}$$

where we have used $\partial_l^2\langle v|c\rangle = \langle\partial_l^2 v|c\rangle + \langle v|\partial_l^2 c\rangle + 2\langle\partial_l v|\partial_l c\rangle = 0$. Substitute these results into Eq.(5.19) we have

$$\begin{aligned}J_i^{(2)} = -e^2(\partial_{x_l}\partial_{x_l}E_j)\int[d\mathbf{k}]\{&\left(\frac{\partial_l V_{vl}-\partial_l V_{cl}}{2(\epsilon_c-\epsilon_v)}+\frac{(V_{vl}+V_{cl})^2}{2(\epsilon_c-\epsilon_v)^2}\right)\mathrm{Im}\langle\partial_i v|\partial_j v\rangle\\ &+\frac{V_{vl}+V_{cl}}{(\epsilon_c-\epsilon_v)^2}[\varepsilon^{ijl}(\vec{V}_v\cdot\Omega_v)-\varepsilon^{ijl}\frac{1}{e}(\nabla\cdot\vec{M}_v))]\\ &+\frac{1}{2}(-2|\langle\partial_l v|c\rangle|^2\,\mathrm{Im}\langle\partial_i v|\partial_j v\rangle+\frac{1}{\epsilon_c-\epsilon_v}\,\mathrm{Im}[\frac{1}{2}\langle\partial_l^2 v|c\rangle\langle\partial_j c|v\rangle(V_{ci}-V_{vi})\\ &\quad+\langle\partial_l v|v\rangle\langle v|\partial_l c\rangle\langle\partial_j c|v\rangle(V_{ci}-V_{vi})-(i\leftrightarrow j)])\}\,.\end{aligned} \tag{5.32}$$

Integral by parts for the first term we have

$$\begin{aligned}J_i^{(2)} = -e^2(\partial_{x_l}\partial_{x_l}E_j)\int[d\mathbf{k}]\{&\frac{2V_{vl}V_{cl}}{(\epsilon_c-\epsilon_v)^2}\,\mathrm{Im}\langle\partial_i v|\partial_j v\rangle-\frac{V_{vl}-V_{cl}}{2(\epsilon_c-\epsilon_v)}\partial_l\,\mathrm{Im}\langle\partial_i v|\partial_j v\rangle\\ &+\frac{V_{vl}+V_{cl}}{(\epsilon_c-\epsilon_v)^2}[\varepsilon^{ijl}(\vec{V}_v\cdot\Omega_v)-\varepsilon^{ijl}\frac{1}{e}(\nabla\cdot\vec{M}_v)]\\ &+\frac{1}{2}(-2|\langle\partial_l v|c\rangle|^2\,\mathrm{Im}\langle\partial_i v|\partial_j v\rangle+\frac{1}{\epsilon_c-\epsilon_v}\,\mathrm{Im}[\frac{1}{2}\langle\partial_l^2 v|c\rangle\langle\partial_j c|v\rangle(V_{ci}-V_{vi})\\ &+\langle\partial_l v|v\rangle\langle v|\partial_l c\rangle\langle\partial_j c|v\rangle(V_{ci}-V_{vi})-(i\leftrightarrow j)])\}\,.\end{aligned} \tag{5.33}$$

Now we make a specific choice of the electric field $\vec{E} = Ee^{iqx}\hat{x}$ and we get current along $\hat{y}$ in a 2D material

$$\begin{aligned}J_y^{(2)} = -\frac{1}{2}e^2(\partial_x^2 E)\int[d^2k]\{&-2|\langle\partial_x v|c\rangle|^2\,\mathrm{Im}\langle\partial_y v|\partial_x v\rangle\\ &+\frac{4V_{vx}V_{cx}}{(\epsilon_c-\epsilon_v)^2}\,\mathrm{Im}\langle\partial_y v|\partial_x v\rangle-\frac{V_{vx}-V_{cx}}{(\epsilon_c-\epsilon_v)}\partial_x\,\mathrm{Im}\langle\partial_y v|\partial_x v\rangle\\ &+\frac{V_{cy}-V_{vy}}{\epsilon_c-\epsilon_v}\,\mathrm{Im}[\frac{1}{2}\langle\partial_x^2 v|c\rangle\langle\partial_x c|v\rangle+\langle\partial_x v|v\rangle\langle v|\partial_x c\rangle\langle\partial_x c|v\rangle]\\ &-\frac{V_{cx}-V_{vx}}{\epsilon_c-\epsilon_v}\,\mathrm{Im}[\frac{1}{2}\langle\partial_x^2 v|c\rangle\langle\partial_y c|v\rangle+\langle\partial_x v|v\rangle\langle v|\partial_x c\rangle\langle\partial_y c|v\rangle])\}\,.\end{aligned} \tag{5.34}$$

The intraband contribution from the first term can be written as

$$\begin{aligned}J_y^{(2)} = -\frac{1}{2}e^2(\partial_x^2 E)\int[d^2k](-2|\langle\partial_x v|c\rangle|^2\,\mathrm{Im}\langle\partial_y v|\partial_x v\rangle)&\\ =-\frac{1}{2}e^2(\partial_x^2 E)\int[d^2k]|\langle\partial_x v|c\rangle|^2\Omega_{yx}&\\ =\frac{1}{2}e^2(\partial_x^2 E)\int[d^2k]\langle\partial_x v|1-P_v|\partial_x v\rangle\Omega_{xy}&\end{aligned} \tag{5.35}$$

where $P_v = |v\rangle\langle v|$ is the projection operator. Note that $\langle\partial_x v|1 - P_v|\partial_x v\rangle$ is the quantum metric tensor. Note that in the above derivation we do not include the diamagnetic current term

$$J_i = -e^2 \int [d^2k] \langle u_n|\partial_i\partial_j H|u_n\rangle A_j \,. \tag{5.36}$$

As we can see it is symmetric and therefore does not contribute to the Hall current.

In the case that we have more than two bands. The current expression will be a bit different. The last term in Eq. (5.18)

$$\begin{aligned}\frac{1}{(\epsilon_m-\epsilon_n)^2}\big(\,\mathrm{Im}[&\frac{1}{2}\langle n|\partial_i H|\partial_l m\rangle\langle\partial_l m|\partial_j H|n\rangle - \langle\partial_l n|\partial_i H|m\rangle\langle\partial_l m|\partial_j H|n\rangle \\ &- \langle\partial_l n|\partial_i H|\partial_l m\rangle\langle m|\partial_j H|n\rangle + \frac{1}{2}\langle\partial_l n|\partial_i H|m\rangle\langle m|\partial_j H|\partial_l n\rangle - (i\leftrightarrow j)] \\ +\frac{1}{2}\,\mathrm{Im}[&\langle n|\partial_i H|\partial_l^2 m\rangle\langle m|\partial_j H|n\rangle + \langle\partial_l^2 n|\partial_i H|m\rangle\langle m|\partial_j H|n\rangle - (i\leftrightarrow j)]\big)\end{aligned} \tag{5.37}$$

will equal to

$$\begin{aligned}\frac{1}{(\epsilon_m-\epsilon_n)^2}\,\mathrm{Im}\Big(&\frac{1}{2}(\epsilon_g-\epsilon_n)(\epsilon_n-\epsilon_k)\langle n|\partial_i g\rangle\langle g|\partial_l m\rangle\langle\partial_l m|k\rangle\langle k|\partial_j n\rangle \\ &(\epsilon_n-\epsilon_k)(\epsilon_m-\epsilon_g)\langle\partial_l n|g\rangle\langle g|\partial_i m\rangle\langle\partial_l m|k\rangle\langle k|\partial_j n\rangle - (i\leftrightarrow j) \\ &(\epsilon_n-\epsilon_m)(\epsilon_k-\epsilon_g)\langle\partial_l n|g\rangle\langle g|\partial_i k\rangle\langle k|\partial_l m\rangle\langle m|\partial_j n\rangle - (i\leftrightarrow j) \\ &\frac{1}{2}(\epsilon_m-\epsilon_g)(\epsilon_m-\epsilon_k)\langle\partial_l n|g\rangle\langle g|\partial_i m\rangle\langle\partial_j m|k\rangle\langle k|\partial_l n\rangle \\ &\frac{1}{2}(\epsilon_n-\epsilon_m)(\epsilon_m-\epsilon_g)\langle\partial_l^2 n|g\rangle\langle g|\partial_i m\rangle\langle m|\partial_j n\rangle (g\neq m) \\ &\frac{1}{2}(\epsilon_n-\epsilon_m)(\epsilon_m-\epsilon_g)\langle g|\partial_l^2 n\rangle\langle\partial_i n|m\rangle\langle\partial_j m|g\rangle (g\neq m) \\ &\frac{1}{2}(\epsilon_n-\epsilon_m)(\epsilon_n-\epsilon_g)\langle\partial_i n|g\rangle\langle g|\partial_l^2 m\rangle\langle m|\partial_j n\rangle (g\neq n) \\ &\frac{1}{2}(\epsilon_n-\epsilon_m)(\epsilon_n-\epsilon_g)\langle\partial_i n|m\rangle\langle\partial_l^2 m|g\rangle\langle g|\partial_j n\rangle (g\neq n)\Big)\,.\end{aligned} \tag{5.38}$$

In the end, we find from Eq. (5.34) that if the system have only two bands and if the bands are flat, the current will be given by a very simple expression

$$J_y^{(2)} = \frac{1}{2}e^2(\partial_x^2 E)\int [d^2k]\langle\partial_x v|1 - P_v|\partial_x v\rangle\Omega_{xy}\,. \tag{5.39}$$

It is an interplay of the Berry curvature and the quantum metric tensor.

5.3 Semiclassical derivation

In this section we would like to derive Eq. (5.39) using a semiclassical approach. As we have mentioned before, in the semiclassical picture, electrons are modeled as wave packets. We

construct a wavepacket centered at x_c and k_c with the eigenstates within a single band.

$$|\Psi\rangle = \int [d^2k]\, a(\vec{k},t)\psi_{\vec{k}}(x,t) \tag{5.40}$$

$$\vec{x}_c = \langle\Psi|\hat{x}|\Psi\rangle \tag{5.41}$$

$$\vec{k}_c = \int [d^2k]\, \vec{k}|a(\vec{k},t)|^2. \tag{5.42}$$

In a static inhomogeneous electric field $\vec{E} = \hat{x}Ee^{iqx} = -\nabla\phi(x)$the local Hamiltonian that such a wave packet feels will be

$$\begin{aligned} H &= H_0(\vec{k}) - e\left[\phi(x_c) + \left.\frac{\mathrm{d}\phi}{\mathrm{d}x}\right|_{x_c}\langle\Psi|(x-x_c)|\Psi\rangle + \frac{1}{2}\left.\frac{\mathrm{d}^2\phi}{\mathrm{d}x^2}\right|_{x_c}\langle\Psi|(x-x_c)^2|\Psi\rangle\right] + \cdots \\ &= H_0(\vec{k}) - e\left(\phi(x_c) - \frac{1}{2}\left.\frac{\mathrm{d}E}{\mathrm{d}x}\right|_{x_c}\langle\Psi|(x-x_c)^2|\Psi\rangle + \cdots\right). \end{aligned} \tag{5.43}$$

We have used the fact that the wave packet is centered at x_c. Since the wavepacket is not a point particle, in the expansion above we have extra terms other than the electrical potential $-e\phi(x_c)$. We work on the first non-zero term

$$\begin{aligned} \langle\Psi|(x-x_c)^2|\Psi\rangle =& \langle\Psi|x^2|\Psi\rangle - x_c^2 \\ =& \int [d^2k]\int [\mathrm{d}^2k']a*(k,t)a(k',t)\langle\psi_{nk}|x^2|\psi_{nk'}\rangle - x_c^2 \\ =& \sum_m \int [\mathrm{d}^2k'']\int [d^2k]\int [\mathrm{d}^2k']a*(k,t)a(k',t)\langle\psi_{nk}|x|\psi_{mk''}\rangle\ \langle\psi_{mk''}|x|\psi_{nk'}\rangle - x_c^2\,. \end{aligned} \tag{5.44}$$

If $m \neq n$ it will be

$$\begin{aligned} &\sum_m \int [\mathrm{d}^2k'']\int [d^2k]\int [\mathrm{d}^2k']a*(k,t)a(k',t)\delta(k''-k')\delta(k-k'')\langle u_{nk}|i\frac{\partial u_{mk}}{\partial k_x}\rangle\langle u_{mk}|i\frac{\partial u_{nk}}{\partial k_x}\rangle \\ &= \int [\mathrm{d}^2k]|a(k,t)|^2\left\langle\frac{\partial u_{nk}}{\partial k_x}\middle|1-P_n\middle|\frac{\partial u_{nk}}{\partial k_x}\right\rangle \\ &= \left.\left\langle\frac{\partial u_{nk}}{\partial k_x}\middle|1-P_n\middle|\frac{\partial u_{nk}}{\partial k_x}\right\rangle\right|_{k=k_c}. \end{aligned} \tag{5.45}$$

If $n = m$ it will be

$$\begin{aligned} &\int [\mathrm{d}^2k'']\int [d^2k]\int [\mathrm{d}^2k']a^*(k,t)a(k',t)\left[\left(i\frac{\partial}{\partial k_x} + \langle u_{nk}|i\frac{\partial u_{nk}}{\partial k_x}\rangle\right)\delta(k-k'')\right] \\ &\qquad\cdot\left[\left(-i\frac{\partial}{\partial k'_x} + \langle u_{nk'}|i\frac{\partial u_{nk'}}{\partial k'_x}\rangle\right)\delta(k'-k'')\right] \\ &= \int [d^2k]\left[\left(-i\frac{\partial}{\partial k_x} + \langle u_{nk}|i\frac{\partial u_{nk}}{\partial k_x}\rangle\right)a^*(k,t)\right]\left[\left(i\frac{\partial}{\partial k_x} + \langle u_{nk}|i\frac{\partial u_{nk}}{\partial k_x}\rangle\right)a(k,t)\right]. \end{aligned} \tag{5.46}$$

Writing the amplitude $a(k,t)$ in the form of $|a(k,t)|e^{-i\gamma(q,t)}$ Eq.(5.46) will be

$$\begin{aligned}\int [d^2k] \left[-ie^{i\gamma}\frac{\partial|a(k,t)|}{\partial k_x} + a^*(k,t)\frac{\partial\gamma}{\partial k_x} + \langle u_{nk}|i\frac{\partial u_{nk}}{\partial k_x}\rangle a^*(k,t)\right] \\ \cdot \left[ie^{-i\gamma}\frac{\partial|a(k,t)|}{\partial k_x} + a(k,t)\frac{\partial\gamma}{\partial k_x} + \langle u_{nk}|i\frac{\partial u_{nk}}{\partial k_x}\rangle a(k,t)\right] \\ = x_c^2 + \int [d^2k] \left(\frac{\partial|a|}{\partial k_x}\right)^2\end{aligned} \tag{5.47}$$

where we have used

$$x_c = \frac{\partial\gamma(k_c,t)}{\partial k_{cx}} + \langle u_{nk}|i\frac{\partial u_{nk}}{\partial k_{cx}}\rangle\,. \tag{5.48}$$

Finally we have

$$\langle\Psi|(x-x_c)^2|\Psi\rangle = \left\langle \frac{\partial u_n}{\partial k_x}\middle|1-P_n\middle|\frac{\partial u_n}{\partial k_x}\right\rangle\Big|_{\vec{k}=\vec{k_c}} + \int [d^2k]\left(\frac{\partial|a|}{\partial k_x}\right)^2 \tag{5.49}$$

The first term is the quantum metric tensor, which describes the intrinsic contribution from the Bloch waves while the second term is related to the details of how the wave packet is constructed. Due to the first term in Eq. (5.49) we will have a correction term to the semiclassical equation for the response to an inhomogeneous electric field. The semiclassical equation will be modified by

$$\dot{\vec{k}}_c = -e\left(E(x_c) + \frac{1}{2}\frac{\mathrm{d}^2E(x_c)}{\mathrm{d}x_c^2}\left\langle\frac{\partial u_n}{\partial k_{cx}}\middle|1-P_n\middle|\frac{\partial u_n}{\partial k_{cx}}\right\rangle\right)\hat{x}\,. \tag{5.50}$$

The Hall velocity we will get from the second term is

$$\delta\dot{\vec{x}}_c = -\delta\dot{\vec{k}}_c\times\vec{\Omega} = e\frac{1}{2}\frac{\mathrm{d}^2E(x_c)}{\mathrm{d}x_c^2}\left\langle\frac{\partial u_n}{\partial k_{cx}}\middle|1-P_n\middle|\frac{\partial u_n}{\partial k_{cx}}\right\rangle\Omega_{xy}\hat{y} \tag{5.51}$$

which leads to the Hall current

$$\delta J_y = e^2\frac{1}{2}\frac{\mathrm{d}^2E}{\mathrm{d}x^2}\int [d^2k]\left\langle\frac{\partial u_n}{\partial k_x}\middle|1-P_n\middle|\frac{\partial u_n}{\partial k_x}\right\rangle\Omega_{xy}\,. \tag{5.52}$$

The result is the same as Eq. (5.39).

There are two comments for the quantum metric tensor and the semiclassical approach:

1. In the Fourier transform we have the relation $\triangle x\cdot\triangle k \geq \frac{1}{2}$. If we make our wavepacket more localized in real space, the width of it in k space will be larger. However, the intrinsic contribution to the Hall conductivity from the Bloch waves u_{nk} is independent of how one construct the wave packet.

2. The quantum metric tensor also appears in construction of the maximally localized Wannier functions [50]. It is the gauge invariant part of the maximally localized Wannier function. For Chern insulators the Wannier function can not be localized in both directions of the two dimensional space. However, we can still have a well defined localization length in one direction.

5.4 Numerical results for Chern insulators

5.4.1 Topological flat band model

First, we study the Hall conductivity in a topological flat band model. There have been several papers studying how to construct 2D lattice tight binding models to produce topological flat bands. Their purpose is to study whether the analytic structure of the wave functions associated with the 2D Landau level is necessary for the fractional quantum Hall state. Here we choose one of the proposed models. It is a two band Chern insulator with two nearly flat bands. The details of this model can be found in [81]. Basically it is modified Haldane model [31] with fine tuned parameters.

We compute the Hall conductivity in the zeroth order and the second order in $\mathbf{q}$. The Hall conductivity in the zeroth order is -3 with $e = 1, h = 1$. For computing the hall conductivity in the second order in $\mathbf{q}$ we use two ways. One is that we use our Eq. (5.39) which is valid for a two band insulator when the bands are flat,

$$J_y^{(2)} = \frac{1}{2} e^2 (\partial_x^2 E) \int [d^2k] \langle \partial_x v | 1 - P_v | \partial_x v \rangle \Omega_{xy} \,. \tag{5.53}$$

This would not be the correct result since there are other terms contributing to the hall conductivity when the bands are not entirely flat. However, it should be very close to the right value since the model we choose has two nearly flat bands. To get the right value we compute it using the expression from Kubo formula which includes all contributions. Indeed, we get the value 0.44 from Eq. (5.39) and 0.43 from the expression including all contributions with $e = 1, h = 1$ and they are very close to each other.

5.4.2 Hofstadter model

In the Hofstadter model, each plaquette in a two dimensional lattice encloses a magnetic flux $\phi = \frac{\phi_0}{n}$ where ϕ_0 is one magnetic flux quantum $\frac{h}{2e}$. The Hofstadter model will converge to continuum Landau levels in the small flux limit $n \to \infty$. Therefore, in the small flux limit we expect to have $C_2 = -\frac{3}{4}$ with the filling factor $\nu = 1$. In this section, we get the numerical results of C_2 for each value of n in a Hofstadter model. In the following we will always work with $\nu = 1$ and we set $e = 1, \hbar = 1$, and the area of each plaquette as 1.

As we can see from Fig.5.1, C_2 is approaching to a constant as n grows. We do linear fitting for C_2 in Fig.5.2 and the linear fitting function is approximately

$$C_2 = -\frac{3}{4} + \frac{2\pi}{8n} \tag{5.54}$$

The constant term is $-\frac{3}{4}$ as expected.

As we have discussed before, C_2 can be divided into two parts, one is related to the Hall viscosity and the other is related to the magnetic susceptibility

$$C_2 = \frac{\eta^A}{\hbar n} - \frac{2\pi}{\nu} \frac{\ell^2}{\hbar \omega_c} B^2 \epsilon''(B) \,. \tag{5.55}$$

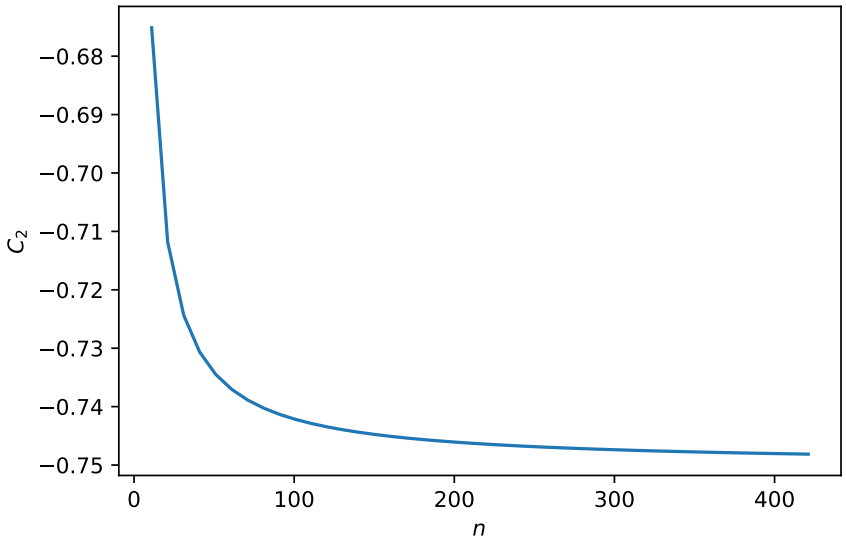

Figure 5.1: The coefficient C_2 of the q^2 term in Hall conductivity for a Hofstadter model with $\frac{1}{n}$ flux quantum in each plaquette.

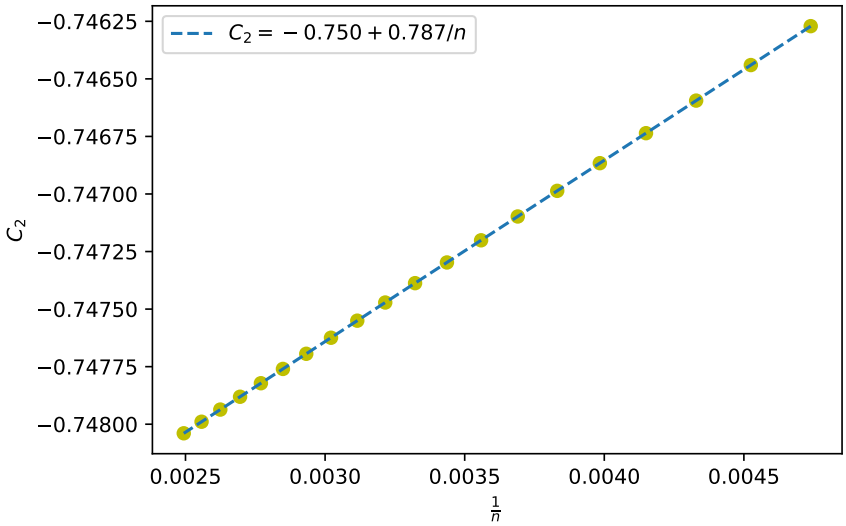

Figure 5.2: Linear fitting for the coefficient C_2 of the q^2 term in Hall conductivity for a Hofstadter model with $\frac{1}{n}$ flux quantum in each plaquette.

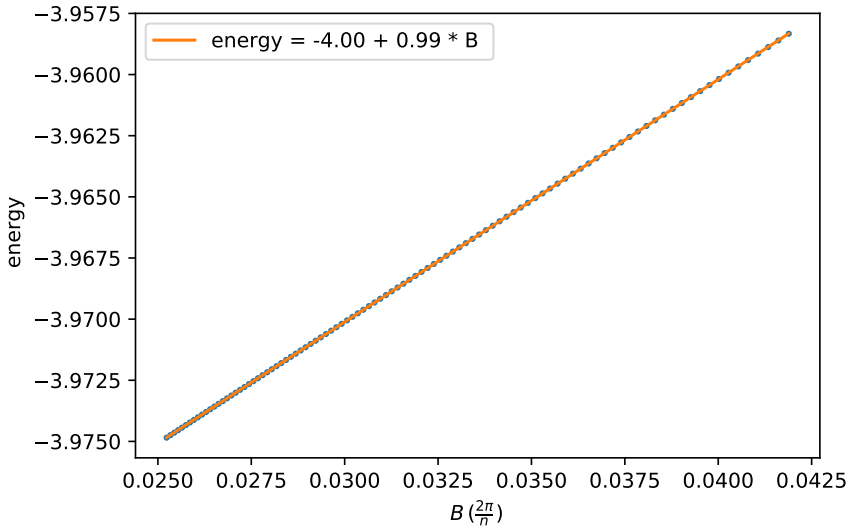

Figure 5.3: The electron energy per unit cell vs the magnetic field B.

If we can get the value of $\epsilon''(B)$ we will have the value of Hall viscosity η^A. There are two ways we can compute $\epsilon''(B)$. One is that we can get it from the electron energy which is a function of the magnetic field. We plot the relation between the one electron energy per unit cell and the magnetic field for the Hofstadter model and do linear fitting. From the fitting result in Fig.5.4.2 we see that the one electron energy per unit cell is linear in the magnetic field in the small flux limit. We thus have

$$\epsilon(B) \cdot 2\pi\ell_B^2 = \epsilon(B) \cdot \frac{2\pi}{B} = B + \text{const} \tag{5.56}$$

and the magnetic susceptibility will be

$$\epsilon''(B) = \frac{2}{2\pi}\,. \tag{5.57}$$

As we know, the magnetic susceptibility of the quantum Hall state at $\nu = 1$ is

$$\epsilon''(B) = \frac{1}{2\pi m}\,. \tag{5.58}$$

By comparing with Eq. (5.57) we can get the mass $m = \frac{1}{2}$. It is because in the small flux limit the energy dispersion is k^2 near $k = 0$.

Another way to compute the magnetic susceptibility is from the symmetric part of the conductivity tensor in the second order in q. The conductivity tensor can be divided into two parts according to Read's paper [15]

$$\sigma^{ij}(q,w) = \frac{1}{m^2 w_c^2}[\frac{i\kappa^{-1}}{w^+} q_\mu q_\nu \epsilon_{\mu i}\epsilon_{\nu j} + (\eta^H(w=0) - \frac{\kappa^{-1}}{w_c})q^2\epsilon_{ij}] \tag{5.59}$$

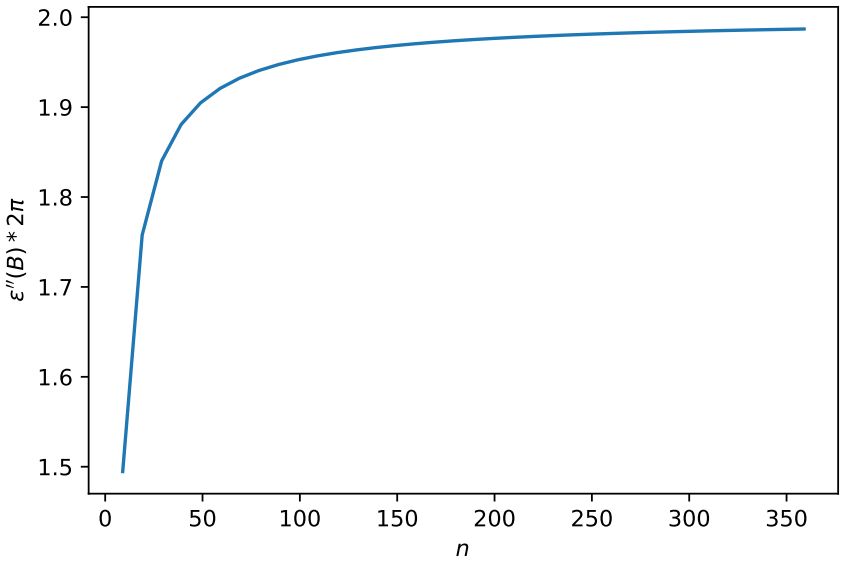

Figure 5.4: Magnetic susceptibility of a Hofstadter model with $\frac{1}{n}$ flux quantum in each plaquette of the 2d lattice.

where

$$\kappa^{-1}(\nu, B) = B^2 \epsilon''(B)\,. \tag{5.60}$$

The first term is symmetric in $i \leftrightarrow j$ and the second term is the hall conductivity tensor. Therefore, we can compute the symmetric part of the conductivity tensor to get the magnetic susceptibility using the Kubo formula

$$\sigma_{ij}^{S} = -\frac{e^2}{iw} \int [d\mathbf{k}] \sum_{n,m} \frac{f(\epsilon_{n,\mathbf{k}-q_x/2}) - f(\epsilon_{m,\mathbf{k}+q_x/2})}{\epsilon_{n,\mathbf{k}-q_x/2} - \epsilon_{m,\mathbf{k}+q_x/2}} \tag{5.61}$$

$$\cdot\, \mathrm{Re}[\langle n_{\mathbf{k}-q_x/2}|\partial_i H|m_{\mathbf{k}+q_x/2}\rangle\langle m_{\mathbf{k}+q_x/2}|\partial_j H|n_{\mathbf{k}-q_x/2}\rangle]\,. \tag{5.62}$$

We plot the values and do linear fitting in Fig.5.5. The fitting function is approximately

$$\epsilon''(B) = \frac{2}{2\pi} - \frac{3}{4n}\,. \tag{5.63}$$

We reach the same conclusion that $\epsilon''(B) = \frac{2}{2\pi}$ and with that we have $\eta^A = \frac{1}{4}$ as expected.

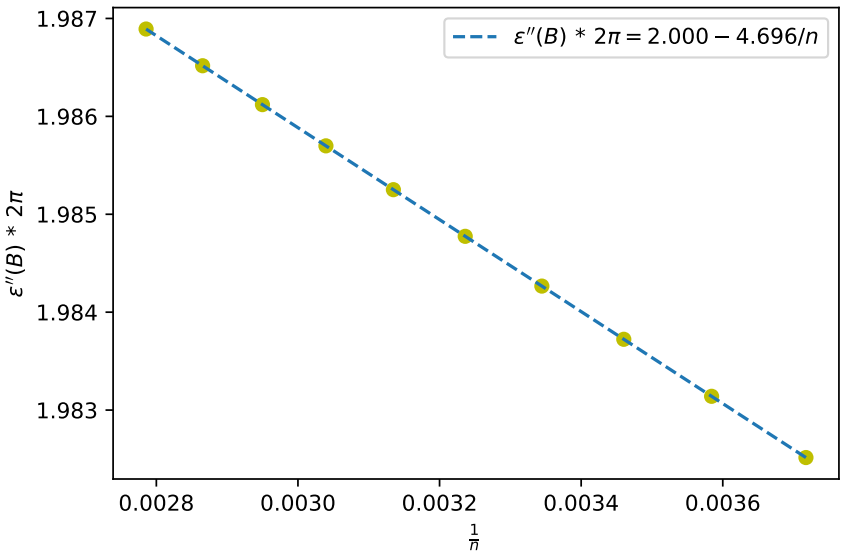

Figure 5.5: Linear fitting for the magnetic susceptibility of a Hofstadter model with $\frac{1}{n}$ flux quantum in each plaquette of the 2d lattice.

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