



DOCTORAL THESIS

**Nonlinear responses in exotic quantum states:
topological superconducting states and
intervalley coherent states**

by:

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Abstract

Quantum condensation yields exotic response phenomena. For example, the Meissner effect, an anomalous electromagnetic phenomenon in superconductors, arises from the condensation of Cooper pairs. The response phenomena have helped us gain deep insights into quantum condensations. In addition, recent developments in nonlinear optics have enabled us to access novel aspects of the exotic quantum states. Nonlinear response measurements can provide rich information about quantum materials, as nonlinear processes can activate degrees of freedom that are inactive in linear processes. In superconductors, the amplitude mode of the order parameter, known as the Higgs mode, gives rise to higher-order optical responses. Nonlinear response phenomena are expected to diversify as they intertwine with the physics of novel quantum states. This thesis focuses on nonlinear response phenomena in topological superconducting states and intervalley coherent states. We numerically demonstrate that characteristic nonlinear responses occur in these exotic quantum states and verify their close ties to intriguing aspects of quantum condensations, such as their topological nature and intervalley coherence.

First, we study the second-order nonlinear optical responses in topological superconductors. We consider s -wave superconductors with Rashba spin-orbit coupling and a magnetic field, a promising platform for topological superconductivity. When the magnetic field is increased, the transition from the topologically trivial to the nontrivial topological superconducting state occurs at the critical magnetic fields. Our numerical results show that the sign of the nonlinear optical conductivity reverses at the topological transition. Furthermore, we analytically discuss the mechanism of the sign reversal. We verify that the band inversion at the topological transition is essential for the sign reversal, and the sign reversal of nonlinear optical conductivity may be universal for topological superconductors. It implies that nonlinear optical measurements can serve as a bulk probe of topological superconductors.

Furthermore, we investigate the effects of collective excitation modes on nonlinear optical responses in topological superconductors. Our main results show that the Higgs modes enhance the nonlinear response when the Fermi level is close to the Dirac point. The enhancement by the Higgs modes does not occur in the single-band s -wave superconductor.

We verify that the multiband character, with split bands due to the Rashba spin-orbit coupling and a Zeeman field, is essential for the enhancement. In particular, the interband pairing is crucial. Recent studies have uncovered that quantum geometry can enhance nonlinear optical responses in topological materials. We show that the peculiar quantum geometry of the Dirac point significantly enhances the nonlinear optical conductivity in the topological superconductor model. We also discuss the interplay of the collective modes and quantum geometry in nonlinear responses.

Next, we focus on the intervalley coherent states as a novel platform for exploring exotic response phenomena. Various types of spin and valley ordering have recently been reported in two-dimensional honeycomb materials. Notably, the intervalley coherent states are attracting attention as exotic states that break valley gauge symmetry. Analogy to superconductivity, which breaks $U(1)$ -gauge symmetry, implies the presence of characteristic response phenomena arising from valley-gauge-symmetry breaking. Here, we focus on valley current generation under surface acoustic waves (SAWs), mechanical vibrations that propagate within the surface of elastic media. The SAWs induce spatio-temporal modulations of the two-dimensional materials placed on the elastic medium. Specifically in honeycomb systems, the spatial modulations are approximately incorporated into the Hamiltonian as an external valley gauge field, namely a pseudogauge field. Thus, the SAWs can drive the spatio-temporally uniform valley currents via nonlinear processes. In this thesis, we demonstrate that the intervalley coherent order leads to anomalous contributions to the valley current driven by SAWs. The anomalous contributions exhibit a characteristic power-law frequency dependence. We furthermore verify that the anomalous valley currents arise from the pseudo-superfluid density, which is analogous to the superfluid density of superconductors. This implies that the anomalous valley currents arise from intrinsic valley-gauge-symmetry breaking and intervalley coherence.

In this thesis, we demonstrate the characteristic nonlinear responses of quantum exotic states through numerical demonstrations and analytical discussions. We present numerical results in figures and discuss the relation between the characteristic behavior and the quantum states. We verify that nonlinear response measurements can be a powerful tool for characterizing these states.

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Publication List

Papers related to the thesis

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Nonlinear optical response in superconductors in magnetic field: Quantum geometry and topological superconductivity,
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Vertex correction for the linear and nonlinear optical responses in superconductors: Multiband effect and topological superconductivity,
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3. Hiroto Tanaka and Youichi Yanase
Surface acoustic wave-driven valley current generation in intervalley coherent states,
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Nonlinear optical response in noncentrosymmetric superconductors,
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Abbreviations

Abbreviation	Definition
BdG	Bogoliubov-de Gennes
CPGE	Circular photogalvanic effect
GL	Ginzburg-Landau
IVC	Intervalley coherent
JDOS	Joint density of states
LPGE	Linear photogalvanic effect
NG mode	Nambu-Goldstone mode
NLE	Nonlinear electric
NRSF	Nonreciprocal superfluid density
R3G	Rhombohedral trilayer graphene
SAW	Surface acoustic wave
SH	Second-harmonic
SHG	Second-harmonic generation
THG	Third-harmonic generation
TSC	Topological superconductor
$\mathcal{P}$ -symmetry	Parity/space-inversion symmetry
$\mathcal{T}$ -symmetry	Time-reversal symmetry
$\mathcal{PT}$ -symmetry	Parity-time-reversal symmetry

Chapter 1

Introduction: overview of nonlinear response phenomena

Response phenomena have revealed remarkable properties of materials and given us deep insights into condensed matter physics. When we apply an external field to a material, it induces changes in some physical quantities within the material. If the external fields are sufficiently small, the induced physical quantities are almost proportional to the external fields. Thus, linear response is one of the most established concepts in response phenomena. Indeed, linear response theory explains macroscopic results in terms of microscopic pictures and accurately describes the relation between external fields and driven physical quantities. However, the linearity breaks down under a strong external field, and the nonlinearity diversifies the response phenomena.

In this chapter, we overview the diversity of the nonlinear response phenomena and their close association with intriguing aspects of quantum states. In Sec. [1.1](#), we introduce the general formula for the response functions that our analysis is based on. In Sec. [1.2](#), we overview the general properties of nonlinear response phenomena and the novel functionality due to the nonlinearity. Furthermore, we focus on the novel nonlinear response phenomena emerging in the quantum materials in Secs. [1.3](#) and [1.4](#). In Sec. [1.3](#), we discuss the relation between the nonlinear response and the quantum geometry and introduce the nonlinear response phenomenon exhibiting a topological nature. In Sec. [1.4](#), we introduce the remarkable nonlinear response phenomena arising from the Cooper pairs' condensation in superconductors. Finally, the organization of this thesis is explained in Sec. [1.5](#).

1.1 Nonlinear response theory

In this section, we introduce linear and nonlinear response functions, which describe the time evolution of the driven physical quantities [1].

We consider a many-body system in thermal equilibrium, described by the Hamiltonian $\hat{\mathcal{H}}$. Without any perturbations, the expectation value of any observable $\hat{A}$ is given by

$$A = \text{Tr} [\hat{\rho}_0 \hat{A}] = \sum P_\lambda \langle \lambda | \hat{A} | \lambda \rangle, \quad (1.1.1)$$

where E_λ is the eigenenergy of the eigenstate $|\lambda\rangle$. P_λ is the canonical distribution function, $P_\lambda = e^{-\beta E_\lambda}/Z$, where Z is the partition function $Z = \sum e^{-\beta E_\lambda}$. The density matrix ρ_0 is defined as $\rho_0 = \sum P_\lambda |\lambda\rangle \langle \lambda|$.

We then introduce a perturbative Hamiltonian coupling to an external field $F_i(t)$,

$$\hat{V}(t) = \sum \hat{B}_i(t) F_i(t) + \sum \hat{C}_{ij}(t) F_i(t) F_j(t) + \cdots, \quad (1.1.2)$$

where the operators are given in the interaction representation $\hat{B}_i(t) = e^{it\mathcal{H}} \hat{B}_i e^{-it\mathcal{H}}$. In the interaction representation, the time evolution of the wave function is described by $|\lambda(t)\rangle = \hat{S}(t, -\infty) |\lambda\rangle$ where the $\hat{S}$ -matrix is defined as

$$\hat{S}(t, -\infty) = T \exp \left(-i \int_{-\infty}^t \hat{V}(t') dt' \right). \quad (1.1.3)$$

The notation T stands for the time-ordering operation. The expectation value of $\hat{A}$ at time t is given by

$$A(t) = \text{Tr} [\hat{\rho}(t) \hat{A}(t)] = \sum P_\lambda \langle \lambda(t) | \hat{A}(t) | \lambda(t) \rangle, \quad (1.1.4)$$

and it is expanded with respect to the external fields,

$$\begin{aligned} A(t) = & \langle \hat{A}(t) \rangle + \sum_i \int_{-\infty}^{\infty} dt_1 \chi_{AB_i}(t_1) F_i(t - t_1) \\ & + \sum_{i,j} \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2 \chi_{AB_i B_j}(t_1, t_2) F_i(t - t_1) F_j(t - t_2) \\ & + \sum_{i,j} \int_{-\infty}^{\infty} dt_1 \chi_{AC_{ij}}(t_1) F_i(t - t_1) F_j(t - t_1) + \cdots. \end{aligned} \quad (1.1.5)$$

The response functions are defined as

$$\chi_{AB_i}(t_1) = i\Theta(t_1) \left\langle \left[\Delta \hat{B}_i(-t_1), \Delta \hat{A}(0) \right] \right\rangle, \quad (1.1.6)$$

$$\chi_{AB_i B_j}(t_1, t_2) = \frac{i^2}{2!} \sum_{\mathcal{P}} \Theta(t_2 - t_1) \Theta(t_1) \left\langle \left[\Delta \hat{B}_j(-t_2), \left[\Delta \hat{B}_i(-t_1), \Delta \hat{A}(0) \right] \right] \right\rangle, \quad (1.1.7)$$

$$\chi_{AC_{ij}}(t_1) = \frac{i}{2!} \sum_{\mathcal{P}} \Theta(t_1) \left\langle \left[\Delta \hat{C}_{ij}(-t_1), \Delta \hat{A}(0) \right] \right\rangle, \quad (1.1.8)$$

where $\Theta(t)$ is the Heaviside step function. The notation $\langle \cdots \rangle$ describes the expectation value under the canonical distribution of an unperturbed Hamiltonian, and $\Delta\hat{X}(t) \equiv \hat{X}(t) - \langle \hat{X}(t) \rangle$. The symbol $\sum_{\mathcal{P}}$ stands for summation over all permutations among the dummy variables, such as (B_i, t_1) and (B_j, t_2) . The second term in Eq. (1.1.5) corresponds to the linear response, while the third and fourth terms describe the second-order nonlinear response. Thus, the linear response function $\chi_{AF_i}(t_1)$ and the second-order nonlinear response function $\chi_{A;F_iF_j}(t_1, t_2)$ are defined as

$$\chi_{AF_i}(t_1) = \chi_{AB_i}(t_1), \quad (1.1.9)$$

$$\chi_{A;F_iF_j}(t_1, t_2) = \chi_{AB_iB_j}(t_1, t_2) + \chi_{AC_{ij}}(t_1)\delta(t_1 - t_2). \quad (1.1.10)$$

Considering the higher-order terms of $F_i(t)$ in the expansion, we can also define the higher-order nonlinear response functions.

Next, we consider the optical response, represented by the linear and nonlinear optical conductivities. We assume the uniform electric field and choose the velocity gauge. The observable of the electric current is defined as

$$\hat{J}^\alpha(\mathbf{A}(t)) = -\frac{\partial \hat{\mathcal{H}}(\mathbf{A}(t))}{\partial A^\alpha(t)}, \quad (1.1.11)$$

where $A^\alpha(t)$ is the vector potential. Because the electric current operator explicitly depends on the vector potential, we also consider the expansion of the current operator with respect to the vector potential. We therefore introduce the higher-order derivatives of the electric current operator as,

$$\hat{J}^{\alpha_1 \cdots \alpha_n} = (-1)^n \left. \frac{\partial^n \hat{\mathcal{H}}(\mathbf{A})}{\partial A^{\alpha_1} \cdots \partial A^{\alpha_n}} \right|_{\mathbf{A}=\mathbf{0}}. \quad (1.1.12)$$

The perturbative part of the Hamiltonian is given by

$$\hat{V}(t) = \sum \hat{J}^\alpha(t) A^\alpha(t) + \sum \hat{J}^{\alpha\beta}(t) A^\alpha(t) A^\beta(t) + \cdots. \quad (1.1.13)$$

The linear and nonlinear response functions are defined as

$$J^\alpha(t) = \sum_{\beta} \int_{-\infty}^{\infty} dt_1 \chi^{\alpha\beta}(t_1) A^\beta(t - t_1) \quad (1.1.14)$$

$$+ \sum_{\beta, \gamma} \int_{-\infty}^{\infty} dt_1 dt_2 \chi^{\alpha; \beta\gamma}(t_1, t_2) A^\beta(t - t_1) A^\gamma(t - t_2) + \cdots. \quad (1.1.15)$$

We can calculate the current response functions $\chi^{\alpha\beta}(t_1)$ and $\chi^{\alpha; \beta\gamma}(t_1, t_2)$ by replacing the operators $\hat{A}$, $\hat{B}$, and $\hat{C}$ in the previous paragraph with $\hat{J}^\alpha$, $\hat{J}^{\alpha\beta}$, and $\hat{J}^{\alpha\beta\gamma}$. After the

Fourier transformation, we can rewrite the linear and nonlinear responses as

$$J^\alpha(\omega) = \sum_{\beta} \chi^{\alpha\beta}(\omega) A^\beta(\omega) \quad (1.1.16)$$

$$+ \sum_{\beta,\gamma} \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} \frac{d\omega_2}{2\pi} \chi^{\alpha;\beta\gamma}(\omega; \omega_1, \omega_2) A^\beta(\omega_1) A^\gamma(\omega_2) 2\pi \delta(\omega - \omega_1 - \omega_2) + \dots \quad (1.1.17)$$

The linear conductivity and the second-order nonlinear conductivity are defined as

$$\sigma^{\alpha\beta}(\omega) = \frac{\chi^{\alpha\beta}(\omega)}{i\omega}, \quad \sigma^{\alpha;\beta\gamma}(\omega; \omega', \omega'') = -\frac{1}{\omega' \omega''} \chi^{\alpha;\beta\gamma}(\omega', \omega''), \quad (1.1.18)$$

and we can obtain the relation between the currents and electric field,

$$J^\alpha(\omega) = \sum_{\beta} \sigma^{\alpha\beta}(\omega) E^\beta(\omega) \quad (1.1.19)$$

$$+ \sum_{\beta,\gamma} \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} \frac{d\omega_2}{2\pi} \sigma^{\alpha;\beta\gamma}(\omega; \omega_1, \omega_2) E^\beta(\omega_1) E^\gamma(\omega_2) 2\pi \delta(\omega - \omega_1 - \omega_2) + \dots \quad (1.1.20)$$

1.2 General properties of nonlinear response

We briefly overview the general properties of nonlinear responses: constraints imposed by symmetry, frequency mixing, and nonreciprocal responses. Furthermore, the nonlinearity of the response phenomena endows materials with novel functionality.

1.2.1 Nonlinear response and symmetry

Response phenomena are closely related to the system's symmetry. Here, we exemplify the second-order nonlinear response,

$$\Delta A = \chi_{A;F_i F_j} F_i F_j. \quad (1.2.1)$$

Under a symmetry operation G such that,

$$G : A \rightarrow \tilde{A}, \quad F_i \rightarrow \tilde{F}_i, \quad \chi_{A;F_i F_j} \rightarrow \tilde{\chi}_{A;F_i F_j}, \quad (1.2.2)$$

the nonlinear response relation is transformed as

$$\Delta \tilde{A} = \tilde{\chi}_{A;F_i F_j} \tilde{F}_i \tilde{F}_j. \quad (1.2.3)$$

When the system is invariant under the operation G , it is known that the nonlinear response function is also invariant, $\chi_{A;F_i F_j} = \tilde{\chi}_{A;F_i F_j}$. This invariance of the response functions is called Neumann's [2]. Thus, we can obtain the relation,

$$\Delta \tilde{A} = \chi_{A;F_i F_j} \tilde{F}_i \tilde{F}_j, \quad (1.2.4)$$

which imposes constraints of the response functions.

For example, the second-order nonlinear optical responses vanishes in space-inversion-symmetric materials. The space-inversion ($\mathcal{P}$) operation changes the electric current and the electric field,

$$\mathcal{P} : J^\alpha \rightarrow -J^\alpha, \quad E^\alpha \rightarrow -E^\alpha. \quad (1.2.5)$$

Thus, we can obtain the relation,

$$J^\alpha = \sigma^{\alpha;\beta\gamma} E^\beta E^\gamma = -J^\alpha, \quad (1.2.6)$$

and the nonlinear conductivity $\sigma^{\alpha;\beta\gamma}$ must be zero in the $\mathcal{P}$ -symmetric materials. Therefore, the second-order nonlinear optical measurements can be a sensitive probe for space inversion parity breaking in materials. Indeed, nonlinear optical techniques have revealed the microscopic $\mathcal{P}$ -symmetry breaking of the antiferromagnets [3] and exotic states in the correlated electron systems [4].

1.2.2 Frequency mixing: second-harmonic generation and photogalvanic effect

Next, we consider the relations between the input and output signal frequencies. In the linear response regime, the input and output signal frequencies are the same when the system is invariant under time translation. Exemplifying the optical responses, the relation is described as

$$J^\alpha(\omega) = \sum_{\beta} \sigma^{\alpha\beta}(\omega) E^\beta(\omega). \quad (1.2.7)$$

On the other hand, nonlinear phenomena can produce an output signal at a new frequency. In the second-order nonlinear responses,

$$J^\alpha(\omega) = \sum_{\beta,\gamma} \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} \frac{d\omega_2}{2\pi} \sigma^{\alpha;\beta\gamma}(\omega; \omega_1, \omega_2) E^\beta(\omega_1) E^\gamma(\omega_2) 2\pi \delta(\omega - \omega_1 - \omega_2), \quad (1.2.8)$$

the nonlinear optical process corresponding to $\sigma^{\alpha;\beta\gamma}(\omega; \omega_1, \omega_2)$ produces the output current with a new frequency $\omega = \omega_1 + \omega_2$ from the two frequency components of the electric field, $E^\beta(\omega_1)$ and $E^\gamma(\omega_2)$. Similarly, the third-order nonlinear responses can outputs the signal with the frequency $\omega = \omega_1 + \omega_2 + \omega_3$ from the three frequency components of the electric field, $E^\beta(\omega_1)$, $E^\gamma(\omega_2)$, and $E^\delta(\omega_3)$.

In particular, the second-order optical responses for a monochromatic light are comprised of the second-harmonic generation (SHG) and the optical rectification, which are denoted by the nonlinear conductivity $\sigma^{\alpha;\beta\gamma}(2\omega; \omega, \omega)$ and $\sigma^{\alpha;\beta\gamma}(0; \omega, -\omega)$ [5]. One class of

optical rectification is the photogalvanic effect, which is the rectified photocurrent arising from a lack of $\mathcal{P}$ -symmetry of the medium [6]. The photogalvanic effect is distinguished from other classes of optical rectification, in which $\mathcal{P}$ -symmetry is broken by inhomogeneity or an external electric field.

SHG, the frequency doubling of light through interaction with media, is a powerful probe of space-inversion symmetry breaking. SHG experiments have revealed microscopic space-inversion-parity-breaking in antiferromagnets [3] and in complex ordered states [4].

The photogalvanic effect is expected to be a new mechanism for solar cells [7]. Interestingly, the intrinsic photogalvanic effect is related to the geometric properties of the Bloch wave functions [6, 7]. Thus, topological materials are potential candidate systems for a new DC photoelectric converter [8].

1.2.3 Nonreciprocal response

Nonlinear responses are closely related to directional responses, namely nonreciprocal responses, characterized by a breakdown of the equivalence between responses driven by right- and left-directed external fields.

We introduce the nonreciprocity and nonreciprocal responses [9, 10]. Onsager's reciprocal theorem relates the time-reversal symmetry of the microscopic dynamics to the change in the macroscopic physical quantity driven by the external field [11]. It says that the linear response function $K_{AB}(q, \omega, B)$ corresponding to the input B and output A with the wavevector q and frequency ω satisfies the relation [12],

$$K_{AB}(q, \omega, B) = \epsilon_A \epsilon_B K_{BA}(-q, \omega, -B), \quad (1.2.9)$$

where $\epsilon_A = \pm 1$ ($\epsilon_B = \pm 1$) specifies the parity of the quantity A (B) for the time-reversal operation. When applied to the linear conductivity, the Onsager reciprocal theorem gives,

$$\sigma^{\alpha\beta}(q, \omega, B) = \sigma^{\beta\alpha}(-q, \omega, -B), \quad (1.2.10)$$

Furthermore, we can obtain the formula for the longitudinal components, $\sigma^{\alpha\alpha}(q, \omega, B) \simeq \sigma^{\alpha\alpha}(\omega) + \kappa q B$. In the transport regime corresponding to the limit $\mathbf{q} \rightarrow \mathbf{0}$, the conductivity is given by $\sigma^{\alpha\beta}(q = 0, \omega, B)$, and it satisfies the relation $\sigma^{\alpha\beta}(\omega, B) = \sigma^{\beta\alpha}(\omega, -B)$. Thus, the linear conductivity does not contain odd-order terms in the magnetic field B .

On the other hand, the nonlinear nonreciprocal transport phenomena, which exhibit the directional I-V characteristics, have the B -linear term. In this case, the resistance is obtained by replacing the wave vector q with the current I , which is suggested by Rikken [13],

$$R = R_0[1 + \beta B^2 + \gamma B I], \quad (1.2.11)$$

where the resistance depends on the direction of the current I , namely unidirectional magnetoresistance [10]. Because Onsager's theorem applies only to linear responses, the replacement of q with the current I is beyond Onsager's theorem. The unidirectional magnetoresistance implies that the nonlinearity of the response can induce novel functionalities that are prohibited in linear responses due to constraints, such as Onsager's nonreciprocal theorem.

1.3 Nonlinear response and quantum geometry

We overview the relation between the nonlinear responses and quantum geometry, the geometric quantities of the Bloch wave functions.

1.3.1 Gauge invariance and quantum geometric quantities

We consider a Hamiltonian $H(\boldsymbol{\lambda})$, which depends on a real parameter $\boldsymbol{\lambda} = (\lambda_1, \dots, \lambda_d)$. The eigenenergy $E_a(\boldsymbol{\lambda})$ and eigenstate $|u_a(\boldsymbol{\lambda})\rangle$ are given by the eigenvalue equation,

$$H(\boldsymbol{\lambda}) |u_a(\boldsymbol{\lambda})\rangle = E_a(\boldsymbol{\lambda}) |u_a(\boldsymbol{\lambda})\rangle. \quad (1.3.1)$$

The gauge transformation, $|u_a(\boldsymbol{\lambda})\rangle \rightarrow |\tilde{u}_a(\boldsymbol{\lambda})\rangle = e^{i\theta_a(\boldsymbol{\lambda})} |u_a(\boldsymbol{\lambda})\rangle$, gives a new vector of the Hilbert space. However, the vector $|\tilde{u}_a(\boldsymbol{\lambda})\rangle$ also satisfies the eigenvalue equation. The gauge degree of freedom of the eigenstates $|u_a(\boldsymbol{\lambda})\rangle$ is just redundancy and does not have physical meaning. Therefore, response functions, which are mathematically described with the eigenenergies and eigenvectors, must be invariant under the gauge transformation. Because, as discussed later, the quantum geometric quantity is invariant under the gauge transformation, it is one of the fundamental factors in the formulas for the response functions.

Next, we introduce gauge-invariant quantities, such as the Berry curvature and the quantum metric. The inner product of two neighboring eigenvectors is given by

$$\langle u_a(\boldsymbol{\lambda}) | u_a(\boldsymbol{\lambda} + d\boldsymbol{\lambda}) \rangle = 1 + \sum_i \langle u_a(\boldsymbol{\lambda}) | \partial_i u_a(\boldsymbol{\lambda}) \rangle d\lambda_i. \quad (1.3.2)$$

The factor $\langle u_a | \partial_i u_a \rangle$ is pure-imaginary and we define the Berry connection $\xi^i(\boldsymbol{\lambda})$ as

$$\xi_a^i(\boldsymbol{\lambda}) = i \langle u_a(\boldsymbol{\lambda}) | \partial_i u_a(\boldsymbol{\lambda}) \rangle. \quad (1.3.3)$$

The Berry connection is dependent on the gauge, and under the gauge transformation $|u_a(\boldsymbol{\lambda})\rangle \rightarrow |\tilde{u}_a(\boldsymbol{\lambda})\rangle = e^{i\theta_a(\boldsymbol{\lambda})} |u_a(\boldsymbol{\lambda})\rangle$ changes,

$$\xi_a^i(\boldsymbol{\lambda}) \rightarrow \xi_a^i(\boldsymbol{\lambda}) - \partial_i \theta_a(\boldsymbol{\lambda}). \quad (1.3.4)$$

The Berry curvature $\Omega_a^{ij}(\boldsymbol{\lambda})$ is defined as

$$\Omega_a^{ij}(\boldsymbol{\lambda}) = \partial_i \xi_a^j(\boldsymbol{\lambda}) - \partial_j \xi_a^i(\boldsymbol{\lambda}), \quad (1.3.5)$$

which is gauge invariant.

One can define the gauge invariant distance of $|u_a(\boldsymbol{\lambda}_1)\rangle$ and $|u_a(\boldsymbol{\lambda}_2)\rangle$,

$$d(|u_a(\boldsymbol{\lambda}_1)\rangle, |u_a(\boldsymbol{\lambda}_2)\rangle) = 1 - |\langle u_a(\boldsymbol{\lambda}_1) | u_a(\boldsymbol{\lambda}_2) \rangle|^2. \quad (1.3.6)$$

In particular, the distance between $|u_a(\boldsymbol{\lambda})\rangle$ and $|u_a(\boldsymbol{\lambda} + d\boldsymbol{\lambda})\rangle$ is given by

$$d(|u_a(\boldsymbol{\lambda})\rangle, |u_a(\boldsymbol{\lambda} + d\boldsymbol{\lambda})\rangle) = \sum_{i,j} g_a^{ij}(\boldsymbol{\lambda}) d\lambda_i d\lambda_j, \quad (1.3.7)$$

where the quantum metric tensor $g_a^{ij}(\boldsymbol{\lambda})$ is defined as

$$g_a^{ij}(\boldsymbol{\lambda}) = \text{Re}[\langle \partial_i u_a(\boldsymbol{\lambda}) | \partial_j u_a(\boldsymbol{\lambda}) \rangle] - \xi_a^i(\boldsymbol{\lambda}) \xi_a^j(\boldsymbol{\lambda}). \quad (1.3.8)$$

Quantum states in crystals are described as the Bloch wave functions $|u_a(\mathbf{k})\rangle$. The quantum geometric quantities in k -space are defined by replacing the parameter $\boldsymbol{\lambda}$ with the Bloch wave vector $\mathbf{k}$.

The quantum geometry of the Bloch wave function, corresponding to the energy band $E_a(\mathbf{k})$, is characterized by the Berry curvature $\Omega_a^{ij}(\mathbf{k})$ and quantum metric $g_a^{ij}(\mathbf{k})$. Furthermore, we introduce interband quantities, which are gauge-invariant because interband transitions are essential for response phenomena. A way to obtain the interband gauge-invariant quantities is to extend the definitions of the quantum geometric quantities to the interband case. The extended Berry connection is defined as

$$\xi_{ab}^i(\mathbf{k}) = i \langle u_a(\mathbf{k}) | \partial_i u_b(\mathbf{k}) \rangle \quad (a \neq b). \quad (1.3.9)$$

The extended Berry connection is not invariant under the gauge transformation and depends on the gauge,

$$\xi_{ab}^i(\mathbf{k}) \rightarrow e^{i(\theta_b(\mathbf{k}) - \theta_a(\mathbf{k}))} \xi_{ab}^i(\mathbf{k}). \quad (1.3.10)$$

One can obtain the gauge invariant quantities by

$$g_{ab}^{ij}(\mathbf{k}) = \text{Re}[\xi_{ab}^i(\mathbf{k}) \xi_{ba}^j(\mathbf{k})], \quad \Omega_{ab}^{ij}(\mathbf{k}) = -2\text{Im}[\xi_{ab}^i(\mathbf{k}) \xi_{ba}^j(\mathbf{k})]. \quad (1.3.11)$$

Another way to get the interband gauge-invariant quantities is to decompose the Berry curvature and the quantum metrics into the interband [14],

$$g_a^{ij}(\mathbf{k}) = \sum_{b \neq a} g_{ab}^{ij}(\mathbf{k}), \quad \Omega_a^{ij}(\mathbf{k}) = \sum_{b \neq a} \Omega_{ab}^{ij}(\mathbf{k}). \quad (1.3.12)$$

Thus, the quantities $g_{ab}^{ij}(\mathbf{k})$ and $\Omega_{ab}^{ij}(\mathbf{k})$ are called the band-resolved quantum metric and Berry curvature [15, 16].

1.3.2 Photogalvanic effect and quantum geometry

Geometric contributions of photogalvanic effect

The photogalvanic effect is characterized the photocurrent conductivity $\sigma^{\alpha\beta\gamma}(0; \omega, -\omega)$. We show the formula of the photocurrent conductivity [16], which contains the quantum geometric quantities. Here, we describe the photocurrent conductivity based on the velocity gauge $\mathbf{E} = -\partial_t \mathbf{A}(t)$.

We consider the photocurrent conductivity in a non-interacting Hamiltonian $H(\mathbf{k})$. With the velocity gauge, the coupling to external electric fields is introduced by the minimal coupling prescription $H(\mathbf{k}) \rightarrow H(\mathbf{k} - e\mathbf{A})$. The perturbative Hamiltonian $\Delta H(\mathbf{k}, t)$ is given by

$$\Delta H(\mathbf{k}, t) \equiv H(\mathbf{k}, \mathbf{A}(t)) - H(\mathbf{k}, \mathbf{0}) \quad (1.3.13)$$

$$= \sum_{n=1} \frac{1}{n!} A^{\alpha_1}(t) \cdots A^{\alpha_n}(t) J^{\alpha_1 \cdots \alpha_n}(\mathbf{k}), \quad (1.3.14)$$

where we introduce the generalized velocity operator

$$J^{\alpha_1 \cdots \alpha_n}(\mathbf{k}) = (-1)^n \left. \frac{\partial^n H(\mathbf{k}, \mathbf{A})}{\partial A^{\alpha_1} \cdots \partial A^{\alpha_n}} \right|_{\mathbf{A}=\mathbf{0}}. \quad (1.3.15)$$

We define the electric current density operator $\mathcal{J}^\alpha(t)$ as

$$\mathcal{J}^\alpha(t) = \frac{1}{2} \sum_{\mathbf{k}} \mathbf{c}_{\mathbf{k}}^\dagger J_{\mathbf{A}}^\alpha(\mathbf{k}, t) \mathbf{c}_{\mathbf{k}}, \quad (1.3.16)$$

where we define $J_{\mathbf{A}}^\alpha(\mathbf{k}, t)$ by

$$J_{\mathbf{A}}^\alpha(\mathbf{k}, t) = \sum_{m=0} \frac{1}{m!} (-1)^m A^{\beta_1}(t) \cdots A^{\beta_m}(t) J^{\alpha\beta_1 \cdots \beta_m}(\mathbf{k}). \quad (1.3.17)$$

The formula for the second-order nonlinear conductivity is obtained as

$$\begin{aligned} & \sigma^{\alpha;\beta\gamma}(\omega; \omega_1, \omega_2) \\ &= \frac{1}{2(i\omega_1 - \eta)(i\omega_2 - \eta)} \sum_{\mathbf{k}} \left[\sum_a \frac{1}{2} J_{aa}^{\alpha\beta\gamma} f_a + \sum_{a,b} \frac{1}{2} \left(\frac{J_{ab}^{\alpha\beta} J_{ba}^\gamma f_{ab}}{\omega_2 + i\eta - E_{ba}} + \frac{J_{ab}^{\alpha\gamma} J_{ba}^\beta f_{ab}}{\omega_1 + i\eta - E_{ba}} \right) \right. \\ & \quad + \sum_{a,b} \frac{1}{2} \frac{J_{ab}^\alpha J_{ba}^{\beta\gamma} f_{ab}}{\omega + 2i\eta - E_{ba}} + \sum_{a,b,c} \frac{1}{2} \frac{J_{ab}^\alpha}{\omega + 2i\eta - E_{ba}} \left(\frac{J_{bc}^\beta J_{ca}^\gamma f_{ac}}{\omega_2 + i\eta - E_{ca}} - \frac{J_{ca}^\beta J_{bc}^\gamma f_{cb}}{\omega_2 + i\eta - E_{bc}} \right) \\ & \quad \left. + \sum_{a,b,c} \frac{1}{2} \frac{J_{ab}^\alpha}{\omega + 2i\eta - E_{ba}} \left(\frac{J_{bc}^\gamma J_{ca}^\beta f_{ac}}{\omega_1 + i\eta - E_{ca}} - \frac{J_{ca}^\gamma J_{bc}^\beta f_{cb}}{\omega_1 + i\eta - E_{bc}} \right) \right], \quad (1.3.18) \end{aligned}$$

where indices a, b, c are spanned by the energy eigenvalues E_a of the unperturbed Hamiltonian $H(\mathbf{k})$ and the energy difference is $E_{ab} \equiv E_a - E_b$. We omit the description

$(\mathbf{k})$ from $J^{\alpha_1 \dots \alpha_n}(\mathbf{k})$ and others. We introduce the Fermi-Dirac distribution function $f_a = (e^{\beta E_a} + 1)^{-1}$ and defined $f_{ab} \equiv f_a - f_b$. The infinitesimal positive parameter η appears due to the adiabatic application of the external fields.

The photogalvanic effect is given by the second-order nonlinear conductivity $\sigma_{\text{PC}}^{\alpha;\beta\gamma}(\omega) = \sigma^{\alpha;\beta\gamma}(0; \omega, -\omega)$. We can decompose the photocurrent conductivities into some contributions,

$$\sigma_{\text{PC}} = \sigma_{\text{D}} + \sigma_{\text{IFS}} + \sigma_{\text{BCD}} + \sigma_{\text{Einj}} + \sigma_{\text{Minj}} + \sigma_{\text{shift}} + \sigma_{\text{gyro}}, \quad (1.3.19)$$

which are termed Drude [17], intrinsic Fermi surface [17], Berry curvature dipole [18], electric injection current [19], magnetic injection current [20], shift current [19, 21], and gyration current [15, 22]. These contributions are described by the gauge invariant formulas [15–17]. The Fermi surface term σ_{FS} , which is allowed in metals, are given by

$$\sigma_{\text{FS}} = \sigma_{\text{D}} + \sigma_{\text{IFS}} + \sigma_{\text{BCD}}. \quad (1.3.20)$$

The formulas have been obtained as

$$\sigma_{\text{D}}^{\alpha;\beta\gamma} = \frac{1}{\Omega^2 + \eta^2} \sum_a [-\partial_{\epsilon_a} \partial_{\beta\gamma} f_a + \partial_{\beta} (\partial_{\gamma} f_a \cdot \partial_{\alpha} \epsilon_a) + \partial_{\gamma} (\partial_{\beta} f_a \cdot \partial_{\alpha} \epsilon_a)], \quad (1.3.21)$$

$$\sigma_{\text{IFS}}^{\alpha;\beta\gamma} = \frac{1}{2} \sum_{a,b} \left[2d_{ba}^{\omega=0} d_{aa}^{-\Omega} \epsilon_{ab} \xi_{ab}^{\alpha} \xi_{ba}^{\beta} \partial_{\gamma} f_{ba} - \xi_{ab}^{\beta} \xi_{ba}^{\gamma} d_{ab}^{\Omega} \partial_{\alpha} f_{ab} \right] + [(\beta, \Omega) \leftrightarrow (\gamma, -\Omega)], \quad (1.3.22)$$

$$\sigma_{\text{BCD}}^{\alpha;\beta\gamma} = -\frac{i}{2\Omega} \sum_a \Omega_a^{\beta\gamma} \partial_{\alpha} f_a, \quad (1.3.23)$$

which have the k -derivative of the Fermi-Dirac distribution. The notation d_{ab}^{ω} is defined as $d_{ab}^{\omega} = (\omega + i\eta - \epsilon_{ab})^{-1}$. On the other hand, the resonant term σ_{res} is written as,

$$\sigma_{\text{Res}} = \sigma_{\text{Einj}} + \sigma_{\text{Minj}} + \sigma_{\text{shift}} + \sigma_{\text{gyro}}. \quad (1.3.24)$$

These contributions are described as

$$\sigma_{\text{Einj}}^{\alpha;\beta\gamma} = -\frac{i\pi}{4\eta} \sum_{a \neq b} (J_{aa}^{\alpha} - J_{bb}^{\alpha}) \Omega_{ba}^{\beta\gamma} f_{ab} \delta(\omega - E_{ba}), \quad (1.3.25)$$

$$\sigma_{\text{Minj}}^{\alpha;\beta\gamma} = -\frac{\pi}{2\eta} \sum_{a \neq b} (J_{aa}^{\alpha} - J_{bb}^{\alpha}) g_{ba}^{\beta\gamma} f_{ab} \delta(\omega - E_{ba}), \quad (1.3.26)$$

$$\sigma_{\text{shift}}^{\alpha;\beta\gamma} = -\frac{\pi}{2} \sum_{a \neq b} \Im \left[[D_{\alpha} \xi^{\beta}]_{ab} \xi_{ba}^{\gamma} + [D_{\alpha} \xi^{\gamma}]_{ab} \xi_{ba}^{\beta} \right] f_{ab} \delta(\omega - E_{ba}), \quad (1.3.27)$$

$$\sigma_{\text{gyro}}^{\alpha;\beta\gamma} = \frac{i\pi}{2} \sum_{a \neq b} \Re \left[[D_{\alpha} \xi^{\beta}]_{ab} \xi_{ba}^{\gamma} - [D_{\alpha} \xi^{\gamma}]_{ab} \xi_{ba}^{\beta} \right] f_{ab} \delta(\omega - E_{ba}), \quad (1.3.28)$$

which have the resonant peak structure given by $\delta(\omega - E_{ba})$. The notation $[D_{\alpha} O]_{ab}$ is defined as

$$[D_{\alpha} O]_{ab} = \partial_{\alpha} O_{ab} - i(\xi_a^{\alpha} - \xi_b^{\alpha}) O_{ab}. \quad (1.3.29)$$

LPGE ($\uparrow$)/ CPGE ($\circ$)	$\mathcal{T}$	$\mathcal{PT}$
($\uparrow$)		Drude*
	Shift current	Magnetic injection current
		Intrinsic Fermi surface*
($\circ$)	Berry curvature dipole*	
	Electric injection current	Gyration current
	Intrinsic Fermi surface*	

Table 1.1: Classification of photocurrent responses in terms of $\mathcal{T}$ and $\mathcal{PT}$ symmetries and of LPGE and CPGE. The entries with the subscript "*" vanish in the absence of the Fermi surface [15].

We discuss the classification of contributions to photogalvanic effects. First, we categorize photogalvanic effects by the polarization of the light that induces photocurrents. The linear photogalvanic effect (LPGE) is the photocurrent generation induced by the linearly polarized light. In contrast, the circular photogalvanic effect (CPGE) gives the difference between photocurrents under left and right circular polarized light. The real and imaginary parts of the photocurrent conductivity, $\text{Re}[\sigma_{\text{PC}}^{\alpha;\beta\gamma}(\omega)]$ and $\text{Im}[\sigma_{\text{PC}}^{\alpha;\beta\gamma}(\omega)]$, corresponds to the LPGE and CPGE, respectively. Furthermore, we classifies the photogalvanic effect in terms of time-reversal ($\mathcal{T}$) and parity-time-reversal ($\mathcal{PT}$) symmetries. The classification is summarized in Table 1.1.

Quantized circular photogalvanic effect in Weyl semimetals

One of the remarkable nonlinear phenomena that exhibits topological nature is the quantized CPGE in three-dimensional Weyl semimetals [23]. Weyl semimetals have even-number degeneracy points in the k -space, Weyl points. The Berry curvature flux Ω_b , which defined as $\Omega_b^\gamma = i\epsilon^{\alpha\beta\gamma} \sum_{a \neq b} \xi_{ba}^\alpha \xi_{ab}^\beta$, diverges from the Weyl points. Interestingly, the surface integral of the Berry curvature flux on the surface around the Weyl points is quantized,

$$C_a = \frac{1}{2\pi} \int_S \Omega_a \cdot d\mathbf{S}, \quad (1.3.30)$$

where C_a is an interger number. Because the C is analogous to the electric charge, C_a is called topological charge.

The topological charge characterizes the CPGE in 3D Weyl semimetals. Here, we rewrite the CPGE current as $\dot{J}^\alpha = \beta^{\alpha\beta} (\mathbf{E} \times \mathbf{E}^*)^\beta$, and the factor $\beta^{\alpha\beta}$ for the electric

injection current is given by

$$\beta^{\alpha\beta}(\omega) = \frac{i\pi e^3}{\hbar^2 V} \sum_{\mathbf{k}} \partial_{\alpha} \epsilon_{12}(\mathbf{k}) \Omega^{\beta}(\mathbf{k}) \delta(\hbar\omega - \epsilon_{12}(\mathbf{k})), \quad (1.3.31)$$

where 1, 2 corresponds to valence and conduction bands, and $\Omega^{\beta}(\mathbf{k}) = \Omega_1^{\beta}(\mathbf{k}) = -\Omega_2^{\beta}(\mathbf{k})$. When the frequency ω is fixed, the delta function select the surface in the k -space where $\hbar\omega = \epsilon_{12}(\mathbf{k})$. The trace of $\beta^{\alpha\beta}$ can be rewritten in terms of the Berry curvature flux,

$$\text{Tr}[\beta] = i \frac{e^3}{2\hbar^2} \int_S \mathbf{\Omega} \cdot d\mathbf{S} = i\pi \frac{e^3}{\hbar^2} C. \quad (1.3.32)$$

In the second equation, we only assume that S is a closed surface encircling the Weyl point, and the quantization does not require the linear or isotropic dispersion.

Although the Weyl point gives rise to a quantized CPGE, the quantization can disappear in the total CPGE. The Nielsen-Ninomiya no-go theorem says that the total topological charge in the whole Brillouin zone must be zero [24]. In mirror symmetric Weyl semimetals, a pair of Weyl points with opposite topological charges is degenerate in energy, leading to an exact cancellation of the $\text{Tr}[\beta]$.

In chiral Weyl semimetals, in which all mirror symmetries are broken, the degeneracy of the Weyl points can be broken. Thus, it is possible that the closed surface S , where $\hbar\omega = \epsilon_{12}(\mathbf{k})$ in the k -space, encircle only one of the Weyl pair. The $\text{Tr}[\beta]$ is quantized in the frequency region $2|\epsilon_L| < \omega < 2|\epsilon_H|$, where ϵ_L and ϵ_H is the energies of the Weyl points. In contrast, the CPGE vanishes outside of the frequency region.

Optical experiments in the chiral Weyl semimetal RhSi verify the prediction for the quantized CPGE [25]. The THz emission spectrometer enables contact-free detection of the LPGE and CPGE currents flowing parallel to the sample surface. The CPGE spectrum shows a rapid drop in amplitude above 0.65 eV. The decrease of the CPGE agrees well with the theoretical prediction [26].

In the relaxation time approximation, the formula of the injection current in the steady state explicitly depends on the relaxation time [15, 16]. Some theoretical works discuss the effects of the impurity scattering and the relaxation on the injection current [26, 27]. When the appropriate frequency difference is set, the quantized CPGE is reproduced even with these relaxation processes.

1.4 Nonlinear response phenomena in superconductors

Superconductors exhibit remarkable electromagnetic properties, such as zero resistance and the Meissner effect. Furthermore, the Cooper pairs' quantum condensation diversifies nonlinear response phenomena. Here, we overview nonlinear responses in superconductors.

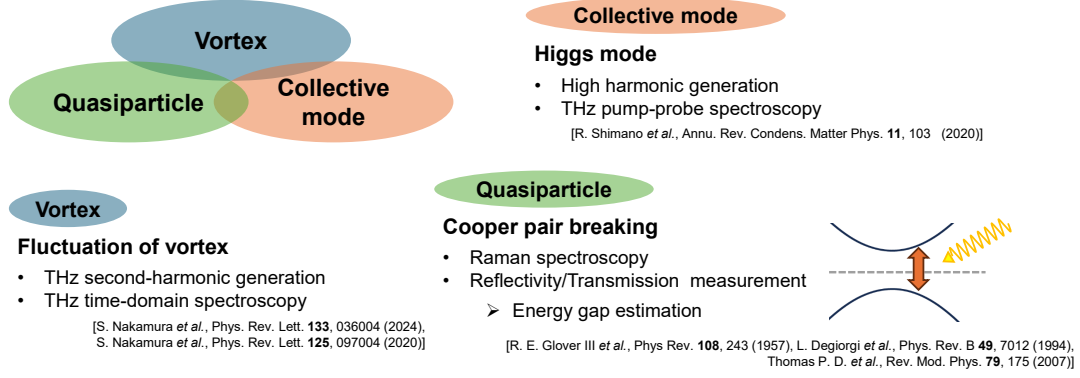


Figure 1.1: Experimental examples of optical responses in superconductors [28–33]. They are explained in terms of Bogoliubov quasiparticles, collective modes, and vortices.

1.4.1 Dynamics of superconductors in optical phenomena

Electron dynamics in superconductors is extremely complicated because superconductivity arises only in many-body systems. However, characteristic phenomena in superconductors has been accurately described in terms of Bogoliubov quasiparticles, collective modes, and vortices [Fig. 1.1]. We introduce experimental examples of optical responses in superconductors where these actors plays important roles.

Superconducting order parameter and Bogoliubov quasiparticle

We show that the effective degrees of freedom in superconducting states are superconducting order parameters and Bogoliubov quasiparticles. Here, we rewrite the partition function in terms of the order parameters and quasiparticles, and explain the relation between these actors [34].

We consider the BCS Hamiltonian,

$$\hat{H}_{\text{BCS}} = \int d^d r c_{\sigma}^{\dagger}(\mathbf{r}) \left[\frac{1}{2m} (-i\nabla - e\mathbf{A})^2 + e\phi - \mu \right] c_{\sigma}(\mathbf{r}) \quad (1.4.1)$$

$$- g \int d^d r c_{\uparrow}^{\dagger}(\mathbf{r}) c_{\downarrow}^{\dagger}(\mathbf{r}) c_{\downarrow}(\mathbf{r}) c_{\uparrow}(\mathbf{r}). \quad (1.4.2)$$

The partition function is obtained by the path integral $Z = \int D(\bar{\psi}, \psi) e^{-S[\bar{\psi}, \psi]}$, and the action S is defined as

$$S[\bar{\psi}, \psi] = \int_0^{\beta} d\tau \int d^d r \left[\bar{\psi}_{\sigma} \left(\partial_{\tau} + ie\phi + \frac{1}{2m} (-i\nabla - e\mathbf{A})^2 - \mu \right) \psi_{\sigma} - g \bar{\psi}_{\uparrow} \bar{\psi}_{\downarrow} \psi_{\downarrow} \psi_{\uparrow} \right], \quad (1.4.3)$$

where ψ_{σ} is a fermionic field. Although the interaction term makes calculations of the partition function difficult, we can decompose the interaction term by the Hubbard-Stratonovich

transformation,

$$\exp \left\{ g \int_0^\beta d\tau \int d^d r \bar{\psi}_\uparrow \bar{\psi}_\downarrow \psi_\downarrow \psi_\uparrow \right\} \quad (1.4.4)$$

$$= \int D(\bar{\Delta}, \Delta) \exp \left\{ - \int_0^\beta d\tau \int d^d r \left[\frac{1}{g} |\Delta|^2 - (\bar{\Delta} \psi_\downarrow \psi_\uparrow + \Delta \bar{\psi}_\uparrow \bar{\psi}_\downarrow) \right] \right\}, \quad (1.4.5)$$

where Δ is a bosonic field. We introduce the Nambu spinor,

$$\bar{\Psi} = (\bar{\psi}_\uparrow, \bar{\psi}_\downarrow), \quad \Psi = \begin{pmatrix} \psi_\uparrow \\ \bar{\psi}_\downarrow \end{pmatrix}. \quad (1.4.6)$$

The partition function is given by

$$Z = \int D(\bar{\psi}, \psi) \int D(\bar{\Delta}, \Delta) \exp \left\{ - \int_0^\beta d\tau \int d^d r \left[\frac{1}{g} |\Delta|^2 - \bar{\Psi} \hat{\mathcal{G}}^{-1} \Psi \right] \right\} \quad (1.4.7)$$

The Gorkov Green function $\hat{\mathcal{G}}^{-1}$ is defined as

$$\hat{\mathcal{G}}^{-1} = \begin{pmatrix} \left[G_0^{(p)} \right]^{-1} & \Delta \\ \bar{\Delta} & \left[G_0^{(h)} \right]^{-1} \end{pmatrix}, \quad (1.4.8)$$

where $\left[G_0^{(p)} \right]^{-1} = -\partial_\tau - \hat{H} + \mu$ and $\left[G_0^{(h)} \right]^{-1} = -\partial_\tau + \hat{H}^\top + \mu$ are Green functions for electrons and holes, respectively ($\hat{H}$ is the non-interacting part of the BCS Hamiltonian).

The partition function is obtained by integrating out the fermionic field ψ_σ and the bosonic field Δ . However, we can approximate the partition function using a fixed bosonic field rather than an integral over bosonic-field paths, namely, the mean-field approximation. When we fix the bosonic field as $\Delta(\tau, \mathbf{r}) = \Delta_0$, the partition function is approximated as

$$Z \simeq Z[\bar{\Delta}_0, \Delta_0] = \int D(\bar{\psi}, \psi) \exp \left\{ - \int_0^\beta d\tau \int d^d r \left[\frac{1}{g} |\Delta_0|^2 - \bar{\Psi} \hat{\mathcal{G}}^{-1}[\bar{\Delta}_0, \Delta_0] \Psi \right] \right\}. \quad (1.4.9)$$

The mean-field Δ_0 is determined by the condition for the saddle point $\frac{\partial Z[\bar{\Delta}_0, \Delta_0]}{\partial \Delta_0} = \frac{\partial Z[\bar{\Delta}_0, \Delta_0]}{\partial \bar{\Delta}_0} = 0$, which is coincide with the gap equation. The effective Hamiltonian for the fermionic field is given by

$$\hat{H}_{\text{eff}} = \begin{pmatrix} \hat{H} & -\Delta \\ -\bar{\Delta} & -\hat{H}^\top \end{pmatrix}, \quad (1.4.10)$$

which is called the Bogoliubov-de Gennes (BdG) Hamiltonian. The BdG Hamiltonian describes the dynamics of Bogoliubov quasiparticles.

Another approximation is expansion of the order parameters. When integrating out the fermionic field, we can obtain the partition function as

$$Z = \int D(\bar{\Delta}, \Delta) \exp \left\{ - \int_0^\beta d\tau \int d^d r \left[\frac{1}{g} |\Delta|^2 - \text{Tr} \ln \hat{\mathcal{G}}^{-1} \right] \right\}. \quad (1.4.11)$$

We consider expansion of $\text{Tr} \ln \hat{\mathcal{G}}^{-1}$ with Δ ,

$$\text{Tr} \ln \hat{\mathcal{G}}^{-1} = \text{Tr} \ln[\hat{\mathcal{G}}_0^{-1}(1 + \hat{\mathcal{G}}_0 \hat{\Delta})] = \text{Tr} \ln \hat{\mathcal{G}}_0^{-1} + \text{Tr} \ln[1 + \hat{\mathcal{G}}_0 \hat{\Delta}] \quad (1.4.12)$$

$$= \text{Tr} \ln \hat{\mathcal{G}}_0^{-1} - \sum_{n=0}^{\infty} \frac{1}{2n} \text{Tr} \left(\hat{\mathcal{G}}_0 \hat{\Delta} \right)^{2n}, \quad (1.4.13)$$

where $\hat{\mathcal{G}}_0 = \hat{\mathcal{G}}|_{\Delta=0}$, and $\hat{\Delta}$ is defined as

$$\hat{\Delta} = \begin{pmatrix} 0 & \Delta \\ \bar{\Delta} & 0 \end{pmatrix}. \quad (1.4.14)$$

Following the expansion, we can obtain the Ginzburg-Landau (GL) action as the effective action for the superconducting order parameters,

$$S_{\text{GL}}[\Delta, \bar{\Delta}] = \beta \int d^d r \left[\frac{r}{2} |\Delta|^2 + \frac{c}{2} |\partial \Delta|^2 + u |\Delta|^4 + \dots \right]. \quad (1.4.15)$$

The coefficients of the GL action are obtained through integrations over higher-order terms of Green functions $\hat{\mathcal{G}}_0$. The GL action has been a powerful tool for the theoretical investigation of the dynamics of superconducting order parameters.

The time evolution of the order parameters and quasiparticles accurately describes the dynamics of superconductors. However, we cannot separate the degrees of freedom of the superconducting state into the order parameters and the quasiparticles completely because these actors are strongly intertwined.

Bogoliubov quasiparticle excitation

Bogoliubov quasiparticle excitation corresponds physically to the breaking of a Cooper pair. The energy bands of the quasiparticle are derived from the diagonalization of the BdG Hamiltonian. In the conventional s -wave superconductors, the superconducting order gaps out the energy bands around the Fermi surface, and the energy gap is given by $\Delta E(\mathbf{k}) \geq 2\Delta$. Thus, the lower energy excitation than 2Δ is prohibited, requiring the linear optical conductivity to be zero for the lower frequency $\omega \leq 2\Delta$. In other words, optical measurements can yield information about the energy gap. Raman spectroscopy [33] and transmission/reflection measurements [31, 32] are established techniques for measuring the superconducting gap.

Collective mode excitations

The collective mode is a coherent motion of many particles, which is characterized by the many-body Green's functions. In ordered states, the collective mode can be interpreted as coherent motions of the order parameters.

The Higgs mode is the amplitude mode of the superconducting order. Although the emergence of the Higgs mode is a universal and fundamental phenomena in superconductors, experimental observation of the Higgs mode in superconductors has been elusive for a long time [30]. One reason is that the Higgs mode has no linear coupling to the external probe, such as electromagnetic field [35]. In other words, the Higgs mode is a charge-neutral scalar excitation.

The first case of observing the Higgs mode was the Raman scattering experiment in a superconductor 2H-NbSe₂, where superconductivity and charge density wave (CDW) coexist [36, 37]. Some experimental and theoretical works revealed the importance of CDW order for the observability of the Higgs mode in the Raman response [38–41]. For decades it was not able to observe the Higgs mode in superconductors without CDW order. After the development of THz optical techniques, it has become possible to observe the Higgs mode clearly in ‘pure’ superconductors, where the long-range order such as CDW order does not exist. THz pump-probe experiments have unveiled the Higgs mode oscillation in quench dynamics [42] and third-harmonic generations (THG) [43–45].

On the other hand, the massless phase mode $\delta\theta$ of the superconducting order parameter $|\Delta|e^{i\theta}$ is termed Nambu-Goldstone (NG) mode. Although the NG mode is also universal in superconductors, the massless collective mode disappears due to the Anderson-Higgs mechanism [46–49]. The NG mode is absorbed into the longitudinal component of the electromagnetic field and is lifted to high energy on the scale of the plasma frequency. Therefore, the remaining low-energy collective excitations are the Higgs mode only in conventional superconductors.

In multiband superconductors, the multiple superconducting order parameters diversify the collective excitation modes. One of the novel collective modes is the Leggett mode, which is the relative phase fluctuation between different bands [50]. Raman spectroscopy observed the Leggett mode in a typical multiband superconductor MgB₂ [51]. Even an uncondensed pairing channel can be relevant. When two Cooper channels are competing, collective excitation of the uncondensed Cooper pairs, known as the Bardasis-Schrieffer mode [52], can occur in optical phenomena. THz nonlinear spectroscopy detected the Bardasis-Schrieffer mode in FeSe [53].

Vortex

Vortices are topological defects of the superconducting order, characterized by the winding number. In noncentrosymmetric superconductors, vortices play an important role in non-reciprocal transport phenomena [54]. Furthermore, recent experiments revealed the motion of the vortex in nonlinear optical phenomena in the THz regime [28, 29]. A THz time-domain spectroscopy shows giant second-harmonic (SH) signals in a thin film of *s*-wave

superconductor NbN, where the $\mathcal{T}$ - and $\mathcal{P}$ -symmetries are broken by injected supercurrents [28]. The temperature dependence of the SH signals is accounted for by the motion of the vortex driven by the THz-electric field in an anharmonic pinning potential. Recently, this THz-SHG detection technique visualizes the picosecond trajectory of vortex motion in a thin film of the iron-based superconductor FeSe_{0.5}Te_{0.5} [29]. It reveals that the vortex core moves independently from the quasiparticles at a significant velocity ≈ 300 km/s.

1.4.2 Unveiled properties of superconductors by nonlinear optics: interplay of nonlinear optical processes and quantum condensations

Recent progress in experimental techniques enables us to access nonlinear optical phenomena in superconductors. The nonlinear optics potentially yield rich information on the degree of freedom inactive in the linear optical responses.

Higgs mode in third-harmonic generation

Let us explain the optical activation of the Higgs mode phenomenologically based on the GL theory. The Higgs mode is a charge-neutral excitation, which has no linear coupling to the electromagnetic field. However, nonlinear optical processes can activate the Higgs mode via a nonlinear light-Higgs coupling.

Here, we denote the superconducting order parameter as $\psi(\mathbf{r}, t)$. Based on the GL theory, the low-energy effective Lagrangian density [35] is given by

$$\mathcal{L} = - \left[a|\psi(\mathbf{r})|^2 + \frac{b}{2}|\psi(\mathbf{r})|^4 + \frac{1}{2m^*}|(-i\nabla - e^*\mathbf{A})\psi(\mathbf{r})|^2 \right] \quad (1.4.16)$$

$$+ c_1\psi(\mathbf{r})^\dagger(i\partial_t - e^*\phi)\psi(\mathbf{r}) + c_2|(i\partial_t - e^*\phi)\psi(\mathbf{r})|^2, \quad (1.4.17)$$

where $\mathbf{A}(\mathbf{r}, t)$ and $\phi(\mathbf{r}, t)$ are the vector and scalar potential of the electromagnetic field. The effective potential is given by the first two terms of Eq. (1.4.16). When the temperature gets lower than the critical temperature ($a < 0$, $b > 0$), the global $U(1)$ symmetry is spontaneously broken, and the order parameter gets down to the bottom of the Mexican-hat-shape potential. We consider the fluctuations of the order parameter from the ground state $\psi(\mathbf{r}) = \psi_0$,

$$\psi(\mathbf{r}) = [\psi_0 + H(\mathbf{r})]e^{i\theta(\mathbf{r})}, \quad (1.4.18)$$

where $H(\mathbf{r})$ and $\theta(\mathbf{r})$ represent amplitude and phase fluctuations, respectively. The Lagrangian is then expanded as

$$\mathcal{L} = 2aH^2 - \frac{1}{2m^*}(\nabla H)^2 - \frac{e^{*2}}{2m^*} \left(\mathbf{A} - \frac{1}{e^*}\nabla\theta \right)^2 (\psi_0 + H)^2 + \dots \quad (1.4.19)$$

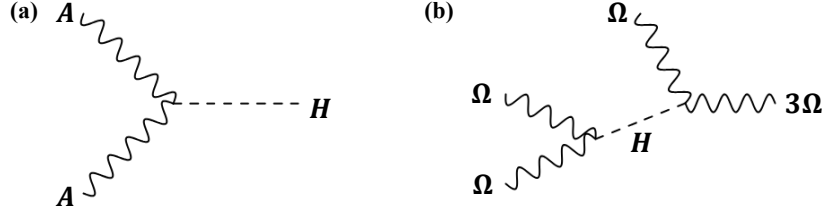


Figure 1.2: (a) Diagrammatic representation of the nonlinear coupling between the Higgs mode and the electromagnetic field. (b) A diagram of the THG mediated by the Higgs mode.

When taking a unitary gauge $\mathbf{A}' = \mathbf{A} - \nabla\theta$, we obtain

$$\mathcal{L} = 2aH^2 - \frac{1}{2m^*}(\nabla H)^2 - \frac{e^{*2}\psi_0^2}{2m^*}\mathbf{A}^2 - \frac{e^{*2}\psi_0}{2m^*}\mathbf{A}^2 H + \dots, \quad (1.4.20)$$

where denoting $\mathbf{A}'$ as $\mathbf{A}$. The mass term of the electromagnetic field appears in the third term. Thus, the photon cannot travel freely inside superconductors but decay exponentially.

We also obtain the light-Higgs nonlinear coupling ($\propto \mathbf{A}^2 H$) in the fourth term. The nonlinear coupling can cause the activation of the Higgs mode by the two-photon processes, which is described schematically in Figure 1.2(a). The Higgs mode can mediate the third-order nonlinear optical processes, such as THGs. The THG spectroscopy shows an enhancement in the spectral amplitude at the frequency $2\omega = 2\psi_0$, corresponding to the excitation energy of the Higgs mode [43, 44].

Nonreciprocal superfluid density

The superfluidity and macroscopic coherence of the superconducting state is characterized by the superfluid density [55]. The superfluid density $\rho^{\alpha\beta}$ is obtained by

$$\rho^{\alpha\beta} = \lim_{\boldsymbol{\lambda} \rightarrow \mathbf{0}} \partial_{\lambda_\alpha} \partial_{\lambda_\beta} F[\mathbf{A} = \boldsymbol{\lambda}], \quad (1.4.21)$$

where $F_{\boldsymbol{\lambda}}$ is the free energy and $\boldsymbol{\lambda}$ is the variational parameters corresponding to the electromagnetic vector potential $\mathbf{A}$. The Meissner effect is also determined by the superfluid density.

When $\mathcal{P}$ and $\mathcal{T}$ symmetries are broken, the nonreciprocity can give rise to response phenomena. Therefore, we can obtain the nonreciprocal correction to the superfluid density, which we refer to as the nonreciprocal superfluid density (NRSF), in $\mathcal{P}$ and $\mathcal{T}$ symmetry broken superconductors [16, 56]. The NRSF is given by

$$f^{\alpha\beta\gamma} = \lim_{\boldsymbol{\lambda} \rightarrow \mathbf{0}} \partial_{\lambda_\alpha} \partial_{\lambda_\beta} \partial_{\lambda_\gamma} F[\mathbf{A} = \boldsymbol{\lambda}]. \quad (1.4.22)$$

The NRSF characterizes the nonreciprocity of the superconducting condensation in view of the electromagnetic responses.

One of the remarkable phenomena induced by the NRSF is the nonreciprocal Meissner effect [56]. The Meissner effect is the screening of magnetic fields by superconductors; magnetic fields can penetrate only a skin depth into a superconductor. The nonreciprocal Meissner effect gives the difference in the penetration depth between whether the magnetic field is parallel or antiparallel to the NRSF vector $\mathbf{f}$, a specific vector given by the NRSF tensor $f^{\alpha\beta\gamma}$. Furthermore, the anomalous optical response arises from the NRSF [16]. In $\mathcal{P}$ - and $\mathcal{T}$ -symmetry-broken superconductors, the second-order nonlinear optical conductivity has an NRSF term,

$$\sigma^{\alpha;\beta\gamma}(\omega_1 + \omega_2, \omega_1, \omega_2) = \frac{f^{\alpha\beta\gamma}}{2\omega_1\omega_2}. \quad (1.4.23)$$

The NRSF term exhibits a characteristic power-law frequency dependence and diverges in the low-frequency limit.

1.5 Organization of this thesis

In this thesis, we show that remarkable response phenomena emerge in exotic quantum states: topological superconducting states and intervalley coherent states. In Chap. 2, we study second-order nonlinear optical responses in topological superconductors. We demonstrate that the direction of photocurrents is inverted at the topological transition. In Chap. 3, we investigate the effects of collective-mode excitations on the nonlinear optical responses in topological superconductors. Our numerical results show that the Higgs mode significantly enhances the nonlinear responses when the Fermi level is close to the Dirac point. In Chap. 4, we verify that anomalous response phenomena analogous to superconductivity arise in valley-gauge-symmetry-broken states, namely, intervalley coherent states. When considering valley-current generations driven by surface acoustic waves (SAWs), valley-gauge-symmetry breaking yields anomalous contributions that exhibit a characteristic power-law dependence on SAW frequency. Finally, the obtained results are summarized in Chap. 5.

Chapter 2

Nonlinear optical responses in topological superconductors

Recent developments in optical techniques have enabled us to access the hidden properties of superconductors [6]. In particular, nonlinear optics has revealed the unique dynamics of superconducting states, such as the Higgs mode in the third-harmonic generations [43–45]. Nonlinear optical techniques can detect the degree of freedom even when it is inactive in the linear optical process. Furthermore, nonlinear optical response is closely related to the topological properties of materials. Quantized circular photogalvanic effect in Weyl semimetals is one of the phenomena that manifests strong ties between nonlinear optics and the topological nature [23].

In this chapter, we focus on the nonlinear optical responses in topological superconductors (TSCs). We numerically demonstrate the photogalvanic effect and second-harmonic generation in *s*-wave superconductors with a Rashba spin-orbit coupling and a Zeeman field, a promising candidate for TSCs [57]. Surprisingly, our numerical results show the sign reversal of the nonlinear optical conductivity at the topological transition. Focusing on the analytical formula of the nonlinear optical responses, we verify that the band inversion is essential for the sign reversal at the topological transition.

This chapter is organized as follows. In Sec. 2.1, we overview candidates for topological superconductivity and introduce the Rashba *s*-wave superconductor model. In Sec. 2.2, we briefly explain the formulation of the nonlinear optical responses in superconductors. The formulas indicate that the joint density of states is essential for the resonant components, and indeed, it helps explain the frequency dependence of the nonlinear responses. We demonstrate photocurrent and second-harmonic generation via numerical calculations and explain their characteristic behavior in terms of the joint density of states. In Sec. 2.3, we show the sign reversal and the specific frequency dependence of the nonlinear optical

conductivity around the topological transition. Finally, we summarize this chapter in Sec. 2.4.

2.1 Topological superconductivity

TSCs have attracted attention from fundamental science to industry. One of the interesting aspects of TSCs is their hosting of Majorana fermions, which are exotic in that they are their own antiparticles [59–61]. Although a superconductor contains electron-electron interactions, the superconducting states are accurately described by an effective single-particle Hamiltonian in which the interaction is approximated as a mean field. The effective Hamiltonian is called the Bogoliubov-de Gennes (BdG) Hamiltonian, which has particle-hole symmetry. Topological states with particle-hole symmetry are classified into BdG symmetry classes as Figure 2.1 [58]. The most common TSC class is class D, where TRS is broken. The class D the 1D p -wave TSC and 2D $p + ip$ TSC. We can make time-reversal-symmetric TSCs, corresponding to class D III, by combining two class D TSCs, with one being the time-reversal version of the other.

The material candidates for topological superconductivity are categorized into natural candidates and artificial candidates [62]. Natural candidates host topological superconductivity on their own, whereas artificial candidates are engineered setups such as heterostructures and nanowire hybrids. A subclass of the natural candidates is spin-triplet pairing superconductors. Although the spin-triplet superconductors are rare in nature, the topological superconductivity can emerge in them without fine-tuning. Heavy fermion superconductors UPt₃, UGe₂, URhGe, UCoGe, and UTe₂ are strong candidates [63].

Artificial structures have also attracted attention as platforms for 1D TSC and 2D TSC. Artificial candidates for 1D TSC are hybrid superconductor-semiconductor nanowire devices under an applied magnetic field [64–67]. Majorana zero modes are expected arise at each end of the wire in such a system. One candidate for 2D TSC is an s -wave SC and

		TRS	PHS	SLS	$d = 1$	$d = 2$	$d = 3$
BdG	D	0	+1	0	$\mathbb{Z}_2$	$\mathbb{Z}$	-
	C	0	-1	0	-	$\mathbb{Z}$	-
	DIII	-1	+1	1	$\mathbb{Z}_2$	$\mathbb{Z}_2$	$\mathbb{Z}$
	CI	+1	-1	1	-	-	$\mathbb{Z}$

Table 2.1: Bogoliubov-de Gennes symmetry classes of single-particle Hamiltonian classified in terms of time-reversal symmetry (TRS), particle-hole symmetry (PHS), and sublattice (or chiral) symmetry (SLS) [58].

a topological insulator. The surface state of the topological insulator is utilized to realize topological superconductivity. Another candidate is artificial heterostructures of an s -wave superconductor and a ferromagnetic insulator [68–70]. In the presence of Rashba spin-orbit coupling and a Zeeman field, the system effectively becomes spinless, allowing the s -wave superconducting state to host Majorana fermions [68, 71, 72].

Much effort has been devoted to the search for topological superconductivity. However, no firm evidence for topological superconductivity has yet been obtained. This is partly because the physical phenomena manifesting the signature of topological superconductivity are limited. Since the existence of the Majorana edge mode is a unique characteristic of topological superconductivity, some theoretical works propose the optical probe for the Majorana edge mode [73–75]. On the other hand, the exploration of bulk probe for topological superconductivity has also been awaited. Therefore, it is highly desirable to elucidate the optical responses that signal the transition to the topological superconducting phase.

2.1.1 Model of Rashba superconductors

In this study, we consider an s -wave superconductor with a Rashba spin-orbit coupling and a Zeeman field, one of the promising artificial candidate for topological superconductivity [68, 71, 72].

Topological superconductivity

We briefly review the topological properties of the Rashba superconductor model [71]. Let us consider the normal Hamiltonian,

$$H_N = H_{\text{kin}} + H_{\text{ASOC}} + H_{\text{Zeeman}}, \quad (2.1.1)$$

$$H_{\text{kin}} = \sum_{\mathbf{k}, s} \xi(\mathbf{k}) C_{\mathbf{k}, s}^\dagger C_{\mathbf{k}, s}, \quad (2.1.2)$$

$$H_{\text{ASOC}} = \sum_{\mathbf{k}, s, s'} \mathbf{g}(\mathbf{k}) \cdot \boldsymbol{\sigma}_{ss'} C_{\mathbf{k}, s}^\dagger C_{\mathbf{k}, s'}, \quad (2.1.3)$$

$$H_{\text{Zeeman}} = \sum_{\mathbf{k}, s, s'} \mathbf{h} \cdot \boldsymbol{\sigma}_{ss'} C_{\mathbf{k}, s}^\dagger C_{\mathbf{k}, s'}, \quad (2.1.4)$$

where $C_{\mathbf{k}, s}$ ($C_{\mathbf{k}, s}^\dagger$) is the annihilation (creation) operator of electrons. The first term H_{kin} is a kinetic energy measured from a chemical potential, which contains the hopping term,

$$\xi(\mathbf{k}) = -2t_1(\cos k_x + \cos k_y) + 4t_2 \cos k_x \cos k_y - \mu. \quad (2.1.5)$$

The second term H_{ASOC} represents a Rashba-type spin-orbit coupling, and the $\mathbf{g}$ -vector is assumed to be

$$\mathbf{g}(\mathbf{k}) = \alpha(\sin k_y, -\sin k_x, 0). \quad (2.1.6)$$

The parameters are set as $t_1 = 1$, $t_2 = 0.2$, and $\alpha = 0.4$ in the numerical calculations. In this study, we consider the Zeeman field H_{Zeeman} only and ignore the orbital effect.

The s -wave superconducting order parameter is phenomenologically introduced as

$$\mathcal{H}_{s\text{-wave}} = \sum_{\mathbf{k}} \left(\psi C_{\mathbf{k}+\mathbf{q},\uparrow}^\dagger C_{-\mathbf{k}+\mathbf{q},\downarrow}^\dagger + \text{H.c.} \right), \quad (2.1.7)$$

where ψ is the magnitude of s -wave superconducting pair potential, and the total momentum of Cooper pairs is $2\mathbf{q}$. The BdG Hamiltonian describes the s -wave superconducting states with $2\mathbf{q} = \mathbf{0}$ in the two-dimensional Rashba system,

$$\begin{aligned} H_{\text{BdG}} &= \frac{1}{2} \sum_{\mathbf{k}} (C_{\mathbf{k}}^\dagger, C_{-\mathbf{k}}^\top) \mathcal{H}(\mathbf{k}) \begin{pmatrix} C_{\mathbf{k}} \\ (C_{-\mathbf{k}}^\dagger)^\top \end{pmatrix} \\ &= \frac{1}{2} \sum_{\mathbf{k}} (C_{\mathbf{k}}^\dagger, C_{-\mathbf{k}}^\top) \begin{pmatrix} \xi(\mathbf{k}) - \mathbf{h} \cdot \boldsymbol{\sigma} + \mathbf{g}(\mathbf{k}) \cdot \boldsymbol{\sigma} & i\psi\sigma_y \\ -i\psi\sigma_y & -\xi(\mathbf{k}) + \mathbf{h} \cdot \boldsymbol{\sigma}^* + \mathbf{g}(\mathbf{k}) \cdot \boldsymbol{\sigma}^* \end{pmatrix} \begin{pmatrix} C_{\mathbf{k}} \\ (C_{-\mathbf{k}}^\dagger)^\top \end{pmatrix}, \end{aligned} \quad (2.1.8)$$

$$(2.1.9)$$

where $C_{\mathbf{k}}^\dagger = (C_{\mathbf{k},\uparrow}^\dagger, C_{\mathbf{k},\downarrow}^\dagger)$. Here, we assume that the Zeeman field is perpendicular to the two-dimensional system. A scenario of topological superconducting states in the Rashba superconductors is based on the following “dual Hamiltonian” $\mathcal{H}^D(\mathbf{k})$,

$$\mathcal{H}^D(\mathbf{k}) = \begin{pmatrix} \psi - h\sigma_z & -i\xi(\mathbf{k})\sigma_y - i\mathbf{g}(\mathbf{k}) \cdot \boldsymbol{\sigma}\sigma_y \\ i\xi(\mathbf{k})\sigma_y + i\mathbf{g}(\mathbf{k}) \cdot \boldsymbol{\sigma}\sigma_y & -\psi + h\sigma_z \end{pmatrix}, \quad (2.1.10)$$

with the unitary transformation

$$\mathcal{H}^D(\mathbf{k}) = D\mathcal{H}(\mathbf{k})D^\dagger, \quad D = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & i\sigma_y \\ i\sigma_y & 1 \end{pmatrix}. \quad (2.1.11)$$

A key feature of the dual Hamiltonian is that the original $\mathbf{g}$ -vector of H_{ASOC} is formally equivalent to the pair potential of the 2D $p + ip$ superconductors. Therefore, the discussion of 2D $p + ip$ TSCs can be used to investigate the topological properties of Rashba superconductors. Ref [71] examine that the topological order emerges when $\xi(\mathbf{k})$, ψ , and h satisfy the condition,

$$\psi^2 + \xi(0,0)^2 < h^2 < \psi^2 + \xi(\pi,0)^2. \quad (2.1.12)$$

The chiral edge mode exists, and non-Abelian statistics of vortices are realized.

In-plane magnetic field and helical superconducting states

In the following, we assume the Zeeman field to be

$$\mathbf{h} = (0, h \cos \theta, h \sin \theta), \quad (2.1.13)$$

with θ being the angle between the magnetic field and the two-dimensional plane. In the Rashba superconductor with an in-plane component of the magnetic field, the total momentum of Cooper pairs gets finite without injecting an electric current [76, 77]. This is called the helical superconducting state, and we introduce the Cooper pairs' momentum $2\mathbf{q}$ in Eq. (2.1.7). The BdG Hamiltonian in the helical superconducting state with $2\mathbf{q}$ is given by

$$H_{\text{BdG}} = \frac{1}{2} \sum_{\mathbf{k}} (c_{\mathbf{k},\mathbf{q}}^\dagger, c_{-\mathbf{k},\mathbf{q}}^\top) \mathcal{H}(\mathbf{k}, \mathbf{q}) \begin{pmatrix} c_{\mathbf{k},\mathbf{q}} \\ (c_{-\mathbf{k},\mathbf{q}}^\dagger)^\top \end{pmatrix} \quad (2.1.14)$$

$$= \frac{1}{2} \sum_{\mathbf{k}} (c_{\mathbf{k},\mathbf{q}}^\dagger, (c_{-\mathbf{k},\mathbf{q}})^\top) \begin{pmatrix} \mathcal{H}_N(\mathbf{k} + \mathbf{q}) & \psi(i\sigma_y) \\ \psi(i\sigma_y)^\dagger & -\mathcal{H}_N(-\mathbf{k} + \mathbf{q})^\top \end{pmatrix} \begin{pmatrix} c_{\mathbf{k},\mathbf{q}} \\ (c_{-\mathbf{k},\mathbf{q}}^\dagger)^\top \end{pmatrix}, \quad (2.1.15)$$

where $c_{\mathbf{k},\mathbf{q}}^\dagger = (c_{\mathbf{k}+\mathbf{q},\uparrow}^\dagger, c_{\mathbf{k}+\mathbf{q},\downarrow}^\dagger)$. The total momentum of Cooper pairs $2\mathbf{q}$ is determined so as to minimize the free energy, which is calculated by

$$F = \langle \mathcal{H} \rangle_{\text{eq}} - TS = -\frac{T}{V} \sum_{\mathbf{k},\alpha} \log \left(1 + e^{-E_\alpha(\mathbf{k}, \mathbf{q})/T} \right). \quad (2.1.16)$$

Note that the minimization is performed with the magnitude of the pair potential ψ fixed in this chapter.

2.2 Photocurrent and second harmonic generation

We investigate the second-order nonlinear optical responses in the Rashba superconductor model. In particular, the photocurrent and second-harmonic generation (SHG) are dominant contributions when the monotonic light is injected.

2.2.1 Formulation

In this chapter, we consider the dynamics of quasiparticles but ignore collective-mode excitations. The dynamics of the quasiparticles are described by the density matrix method based on the BdG Hamiltonian [16]. We then obtain the formula for the nonlinear optical conductivity in the superconducting state using the density-matrix method. Note that we focus on the second-order nonlinear optical responses in clean superconductors [16, 57, 78–83], which contrast with those induced by impurity scattering [84, 85].

We briefly explain the formulation of the nonlinear conductivity. With the velocity gauge $\mathbf{E} = -\partial_t \mathbf{A}$, the coupling to external electric fields is introduced by the minimal coupling description $\mathcal{H}_N(\mathbf{k}) \rightarrow \mathcal{H}_N(\mathbf{k} - e\mathbf{A})$. Thus, the vector potential $\mathbf{A}$ dependence of

the Hamiltonian is expressed as

$$\mathcal{H}(\mathbf{k}, \mathbf{q}, \mathbf{A}) = \begin{pmatrix} \mathcal{H}_N(\mathbf{k} + \mathbf{q} - e\mathbf{A}) & \psi(i\sigma_y) \\ \psi(i\sigma_y)^\dagger & -\mathcal{H}_N(-\mathbf{k} + \mathbf{q} - e\mathbf{A})^\top \end{pmatrix}. \quad (2.2.1)$$

The perturbative Hamiltonian $\Delta\mathcal{H}(\mathbf{k}, \mathbf{q}, t)$ is given by

$$\Delta\mathcal{H}(\mathbf{k}, \mathbf{q}, t) \equiv \mathcal{H}(\mathbf{k}, \mathbf{q}, \mathbf{A}(t)) - \mathcal{H}(\mathbf{k}, \mathbf{q}, \mathbf{0}) = \sum_{n=1} \frac{1}{n!} A^{\alpha_1}(t) \cdots A^{\alpha_n}(t) \mathcal{J}^{\alpha_1 \cdots \alpha_n}(\mathbf{k}, \mathbf{q}), \quad (2.2.2)$$

where we introduce the generalized velocity operator

$$\mathcal{J}^{\alpha_1 \cdots \alpha_n}(\mathbf{k}, \mathbf{q}) = (-1)^n \left. \frac{\partial^n \mathcal{H}(\mathbf{k}, \mathbf{q}, \mathbf{A})}{\partial A^{\alpha_1} \cdots \partial A^{\alpha_n}} \right|_{\mathbf{A}=\mathbf{0}}. \quad (2.2.3)$$

We define the electric current density operator $J^\alpha(t)$

$$J^\alpha(t) = \frac{1}{2} \sum_{\mathbf{k}} \mathbf{c}_{\mathbf{k}, \mathbf{q}}^\dagger \mathcal{J}_A^\alpha(\mathbf{k}, \mathbf{q}, t) \mathbf{c}_{\mathbf{k}, \mathbf{q}}, \quad (2.2.4)$$

where we define $\mathcal{J}_A^\alpha(\mathbf{k}, \mathbf{q}, t)$ by

$$\mathcal{J}_A^\alpha(\mathbf{k}, \mathbf{q}, t) = \sum_{m=0} \frac{1}{m!} (-1)^m A^{\beta_1}(t) \cdots A^{\beta_m}(t) \mathcal{J}^{\alpha\beta_1 \cdots \beta_m}(\mathbf{k}, \mathbf{q}). \quad (2.2.5)$$

The formula for the second-order nonlinear conductivity is obtained as

$$\begin{aligned} & \sigma^{\alpha; \beta\gamma}(\omega; \omega_1, \omega_2) \\ &= \frac{1}{2(i\omega_1 - \eta)(i\omega_2 - \eta)} \sum_{\mathbf{k}} \left[\sum_a \frac{1}{2} J_{aa}^{\alpha\beta\gamma} f_a + \sum_{a,b} \frac{1}{2} \left(\frac{J_{ab}^{\alpha\beta} J_{ba}^\gamma f_{ab}}{\omega_2 + i\eta - E_{ba}} + \frac{J_{ab}^{\alpha\gamma} J_{ba}^\beta f_{ab}}{\omega_1 + i\eta - E_{ba}} \right) \right. \\ & \quad + \sum_{a,b} \frac{1}{2} \frac{J_{ab}^\alpha J_{ba}^{\beta\gamma} f_{ab}}{\omega + 2i\eta - E_{ba}} + \sum_{a,b,c} \frac{1}{2} \frac{J_{ab}^\alpha}{\omega + 2i\eta - E_{ba}} \left(\frac{J_{bc}^\beta J_{ca}^\gamma f_{ac}}{\omega_2 + i\eta - E_{ca}} - \frac{J_{ca}^\beta J_{bc}^\gamma f_{cb}}{\omega_2 + i\eta - E_{bc}} \right) \\ & \quad \left. + \sum_{a,b,c} \frac{1}{2} \frac{J_{ab}^\alpha}{\omega + 2i\eta - E_{ba}} \left(\frac{J_{bc}^\gamma J_{ca}^\beta f_{ac}}{\omega_1 + i\eta - E_{ca}} - \frac{J_{ca}^\gamma J_{bc}^\beta f_{cb}}{\omega_1 + i\eta - E_{bc}} \right) \right], \quad (2.2.6) \end{aligned}$$

where indices a, b, c are spanned by the energy eigenvalues E_a of the unperturbed Hamiltonian $\mathcal{H}(\mathbf{k}, \mathbf{q})$ and the energy difference is $E_{ab} = E_a - E_b$. The matrix elements of the current operator $\mathcal{J}^{\alpha_1 \cdots \alpha_n}(\mathbf{k}, \mathbf{q})$ is described as $J_{ab}^{\alpha_1 \cdots \alpha_n}$ on the band basis. We omit the description $(\mathbf{k}, \mathbf{q})$ from $\mathcal{J}^{\alpha_1 \cdots \alpha_n}(\mathbf{k}, \mathbf{q})$ and others. We introduce the Fermi-Dirac distribution function $f_a = (e^{\beta E_a} + 1)^{-1}$ and defined $f_{ab} = f_a - f_b$. The infinitesimal positive parameter η appears due to the adiabatic application of the external fields.

Generalized Berry connection

Here, we focus on the photocurrent conductivity $\sigma^{\alpha; \beta\gamma}(0, \omega, -\omega)$. The photocurrent conductivity in superconducting states can be rewritten as the explicitly gauge-invariant formula, as that in the normal states in Sec.1.3.2. To obtain the gauge-invariant formula, we introduce the generalized Berry connection.

Before discussion of the generalized Berry connection in superconducting states, we review relation between the $\mathbf{k}$ -derivative Berry connection and the photocurrent conductivity in the normal states. The general formula of the second-order nonlinear conductivity is described in terms of the matrix elements of the current operators as Eq. (1.3.18). With the Hellmann-Feynman theorem, the paramagnetic current operator is obtained as

$$J_{ab}^\alpha(\mathbf{k}) = \frac{\partial}{\partial k_\alpha} E_a \delta_{ab} + i E_{ab} \xi_{ab}^\alpha(\mathbf{k}), \quad (2.2.8)$$

with the $\mathbf{k}$ -derivative Berry connection

$$\xi_{ab}^\alpha(\mathbf{k}) = i \left\langle u_a(\mathbf{k}) \left| \frac{\partial u_b(\mathbf{k})}{\partial k_\alpha} \right. \right\rangle, \quad (2.2.9)$$

defined with the periodic part of the Bloch states $\{|u_a(\mathbf{k})\rangle\}$. The relation between the Berry connection and the paramagnetic current operator originates from the equivalence of differentiation with respect to the vector potential $\mathbf{A}$ and with respect to the momentum $-\mathbf{k}$, which is justified by the minimal coupling $\mathbf{p} \rightarrow \mathbf{p} - e\mathbf{A}$. This relation is essential for rewriting the formula explicitly gauge-invariant.

In contrast to the normal state, the Hellmann-Feynman relation in Eq.(2.2.8) fails in the superconducting state. In the BdG formalism, particles with opposite charges, that is, electron and hole, are treated on equal footing. Thus, a naive treatment based on the minimal coupling $\mathbf{p} \rightarrow \mathbf{p} - e\mathbf{A}$ is not justified. Therefore, we consider $\boldsymbol{\lambda}$ -derivative Berry connections instead of the $\mathbf{k}$ -derivative ones, where $\boldsymbol{\lambda}$ is the vector potential parameter.

We introduce the variational parameter $\boldsymbol{\lambda}$ with bearing the minimal coupling in mind,

$$\mathcal{H}(\mathbf{k}, \mathbf{q}, \mathbf{A} = \mathbf{0}) \rightarrow \mathcal{H}_\lambda(\mathbf{k}, \mathbf{q}) = \mathcal{H}(\mathbf{k}, \mathbf{q}, \mathbf{A} = \boldsymbol{\lambda}). \quad (2.2.10)$$

Since the vector parameter $\boldsymbol{\lambda}$ plays the same role as the (spatially uniform and time-independent) vector potential, we obtain the relation

$$J_{ab}^\alpha(\mathbf{k}) = - \lim_{\boldsymbol{\lambda} \rightarrow \mathbf{0}} \left\langle a_\lambda \left| \frac{\partial \mathcal{H}_\lambda}{\partial \lambda_\alpha} \right| b_\lambda \right\rangle, \quad (2.2.11)$$

where $|a_\lambda(\mathbf{k}, \mathbf{q})\rangle$ is the eigenstate of the Hamiltonian $\mathcal{H}_\lambda(\mathbf{k}, \mathbf{q})$. The Hellmann-Feynman relation for the vector potential parametrization is obtained from

$$\left\langle a_\lambda \left| \frac{\partial \mathcal{H}_\lambda}{\partial \lambda_\alpha} \right| b_\lambda \right\rangle = \frac{\partial E_a(\boldsymbol{\lambda})}{\partial \lambda_\alpha} \delta_{ab} - i [\xi^{\lambda_\alpha}, H_\lambda]_{ab}, \quad (2.2.12)$$

where we defined the connection ξ^{λ_α} with the replacement $\partial_{k_\alpha} \rightarrow \partial_{\lambda_\alpha}$ in Eq. (2.2.9):

$$\xi_{ab}^{\lambda_\alpha} = i \left\langle a_\lambda \left| \frac{\partial b_\lambda}{\partial \lambda_\alpha} \right. \right\rangle. \quad (2.2.13)$$

The relation between the paramagnetic current operator and Berry connection is as follows:

$$J_{ab}^\alpha = \lim_{\boldsymbol{\lambda} \rightarrow \mathbf{0}} \left[- \frac{\partial E_a(\boldsymbol{\lambda})}{\partial \lambda_\alpha} \delta_{ab} - i E_{ab}(\boldsymbol{\lambda}) \xi_{ab}^{\lambda_\alpha} \right]. \quad (2.2.14)$$

This relation plays an essential role in simplifying the optical conductivity formulas. The linear optical conductivity and photocurrent response have been decomposed into normal and anomalous contributions with using the vector potential parametrization [16].

Normal and anomalous photocurrent responses

In the gapful superconductors at low temperatures, the total photocurrent conductivity is decomposed into the two components

$$\sigma = \sigma_n + \sigma_a. \quad (2.2.15)$$

The former is a normal photocurrent which is a counterpart of the photocurrent in the normal state, while the latter is an anomalous photocurrent unique to the superconducting state [16].

The normal photocurrent consists of the four contributions

$$\sigma_n = \sigma_{\text{Einj}} + \sigma_{\text{Minj}} + \sigma_{\text{shift}} + \sigma_{\text{gyro}}, \quad (2.2.16)$$

where the terminology is same as Eq (1.3.19). The formulas based on the BdG Hamiltonian are obtained as

$$\sigma_{\text{Einj}}^{\alpha;\beta\gamma} = -\frac{i\pi}{8\eta} \sum_{a \neq b} (J_{aa}^\alpha - J_{bb}^\alpha) \Omega_{ba}^{\lambda_\beta \lambda_\gamma} f_{ab} \delta(\omega - E_{ba}), \quad (2.2.17)$$

$$\sigma_{\text{Minj}}^{\alpha;\beta\gamma} = \frac{\pi}{4\eta} \sum_{a \neq b} (J_{aa}^\alpha - J_{bb}^\alpha) g_{ba}^{\lambda_\beta \lambda_\gamma} f_{ab} \delta(\omega - E_{ba}), \quad (2.2.18)$$

$$\sigma_{\text{shift}}^{\alpha;\beta\gamma} = -\frac{\pi}{4} \sum_{a \neq b} \Im \left[\left[D_{\lambda_\alpha} \xi^{\lambda_\beta} \right]_{ab} \xi_{ba}^{\lambda_\gamma} + \left[D_{\lambda_\alpha} \xi^{\lambda_\gamma} \right]_{ab} \xi_{ba}^{\lambda_\beta} \right] f_{ab} \delta(\omega - E_{ba}), \quad (2.2.19)$$

$$\sigma_{\text{gyro}}^{\alpha;\beta\gamma} = \frac{i\pi}{4} \sum_{a \neq b} \Re \left[\left[D_{\lambda_\alpha} \xi^{\lambda_\beta} \right]_{ab} \xi_{ba}^{\lambda_\gamma} - \left[D_{\lambda_\alpha} \xi^{\lambda_\gamma} \right]_{ab} \xi_{ba}^{\lambda_\beta} \right] f_{ab} \delta(\omega - E_{ba}). \quad (2.2.20)$$

WE defined the geometric quantities such as the Berry curvature ($\Omega_{ab}^{\lambda_\alpha \lambda_\beta}$) and quantum metric ($g_{ab}^{\lambda_\alpha \lambda_\beta}$), and the covariant derivative (D_{λ_α}). The covariant derivative is associated with the $U(N)$ -Berry connection $\mathcal{A}^\alpha$ of N -fold energy-degenerate bands. When the pair potential is zero, we can easily reproduce the photocurrent conductivity in the normal state by replacing λ with $\mathbf{k}$ in the normal photocurrent.

The microscopic mechanism is explained based on the illustration of Fig. 2.1. In the superconducting state, the type A and type B transitions can contribute to the normal photocurrents [Fig. 2.1(b)]. The type A transition is similar to the interband transition in the normal state shown in Fig. 2.1(a), while the type B transition is unique to the superconducting states. The contributions arising in the superconducting states exhibit the frequency dependence characteristic of a type B transition.

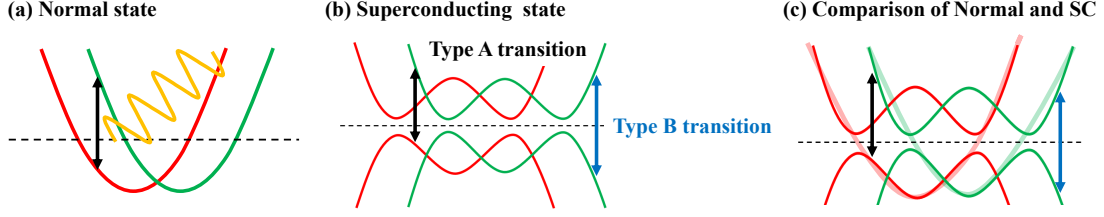


Figure 2.1: Schematic illustration of the transitions that contribute to the second-order nonlinear responses. Note that the Zeeman field is set to be zero in the illustration for simplicity. (a) In the normal state, the interband transition dominantly contributes to the second-order nonlinear response. (b) In the superconducting state, the transition processes that make dominant contributions are categorized into type A and type B transitions. Here, we define the eigenenergy of the quasiparticles $E_i(\mathbf{k})$ as $E_1(\mathbf{k}) \leq E_2(\mathbf{k}) \leq 0 \leq E_3(\mathbf{k}) \leq E_4(\mathbf{k})$. Type A corresponds to $E_1 \leftrightarrow E_4$ and $E_2 \leftrightarrow E_3$ transitions, type B is $E_1 \leftrightarrow E_3$ and $E_2 \leftrightarrow E_4$ transitions. (c) A Type A transition is similar to an interband transition in the normal state. Thus, the contributions of the nonlinear responses in the normal states appear even superconducting via the type A transition. On the other hand, the type B transition makes unique contributions to the superconducting states.

Because of the nonequivalence of λ and $\mathbf{k}$, the anomalous conductivity appears in the superconducting state. The anomalous photocurrent consists of two contributions

$$\sigma_a = \sigma_{\text{NRSF}} + \sigma_{\text{CD}}, \quad (2.2.21)$$

which are termed nonreciprocal superfluid density (NRSF) term, and conductivity derivative term, respectively. The formulas are given by

$$\sigma_{\text{NRSF}}^{\alpha;\beta\gamma} = \lim_{\lambda \rightarrow 0} -\frac{1}{2\omega^2} \partial_{\lambda_\alpha} \partial_{\lambda_\beta} \partial_{\lambda_\gamma} F_\lambda, \quad (2.2.22)$$

$$\sigma_{\text{CD}}^{\alpha;\beta\gamma} = \lim_{\lambda \rightarrow 0} \frac{1}{4\omega^2} \partial_{\lambda_\alpha} \left[\sum_{a \neq b} J_{ab}^\beta J_{ba}^\gamma f_{ab} \left(\frac{1}{\omega - E_{ab}} + \frac{1}{E_{ab}} \right) \right]. \quad (2.2.23)$$

In Eq. (2.2.22), F_λ is the free energy of the BdG Hamiltonian.

A unique property of nonlinear optical responses of superconductors is the low-frequency divergence. Indeed, the anomalous photocurrent shows divergent behaviors in the low-frequency regime. We already discussed the NRSF term in Sec. 1.4.2. The NRSF shows the ω^{-2} divergence at the low frequency. The conductivity derivative term in the low-frequency limit ($\omega \rightarrow 0$) is reduced to the λ -derivative of the total Berry curvature $B_d^{\alpha\delta}$:

$$\sigma_{\text{CD}}^{\alpha;\beta\gamma} \rightarrow \sigma_{\text{sCD}}^{\alpha;\beta\gamma} = \lim_{\lambda \rightarrow 0} \frac{i}{4\Omega} \epsilon_{\beta\gamma\delta} \partial_{\lambda_\alpha} \left(\sum_a \Omega_a^{\lambda_\delta} f_a \right) = \frac{i}{4\Omega} \epsilon_{\beta\gamma\delta} B_d^{\alpha\delta}. \quad (2.2.24)$$

The conductivity derivative term in the static limit $\sigma_{\text{sCD}}^{\alpha;\beta\gamma}$ is represented by Berry curvature

LPGE ($\Downarrow$)/ CPGE ($\Uparrow$)	$\mathcal{T}$	$\mathcal{PT}$
($\Downarrow$)	Shift current	Magnetic injection current NRSF*
($\Uparrow$)	Electric injection current Conductivity derivative*	Gyration current

Table 2.2: Classification of photocurrent responses in gapful superconductors in terms of $\mathcal{T}$ and $\mathcal{PT}$ symmetries and of linear photogalvanic effect (LPGE) and circular photogalvanic effect (CPGE). The entries with the superscript, *, vanish in the normal states.

derivative $B_d^{\alpha\delta}$ and shows the ω^{-1} divergence. The diverging behaviors are absent in normal metals and insulators and unique to superconductors.

We classify the contributions of photocurrents in the gapful superconductors in terms of $\mathcal{T}$ - and $\mathcal{PT}$ - symmetries and of linear photogalvanic effect (LPGE) and circular photogalvanic effect (CPGE). For the anomalous photocurrent, the NRSF term is prohibited in the $\mathcal{T}$ -symmetric system, while the conductivity derivative term disappears in the $\mathcal{PT}$ -symmetric system. The classification is summarized in Table 2.2.

Anomalous contribution to general nonlinear responses

The low-frequency divergent behavior also emerges in other nonlinear responses such as the SHG. The anomalous parts of nonlinear responses have been obtained by the Green function method [16]. The low-frequency anomalous contribution reads as

$$\sigma_a^{\alpha;\beta\gamma}(\omega_1 + \omega_2; \omega_1, \omega_2) = \sigma_{\text{NRSF}}^{\alpha;\beta\gamma}(\omega_1 + \omega_2; \omega_1, \omega_2) + \sigma_{\text{CD}}^{\alpha;\beta\gamma}(\omega_1 + \omega_2; \omega_1, \omega_2), \quad (2.2.25)$$

where $O(\omega_1^a \omega_2^b)$ terms are suppressed ($a + b \geq 0$). In gapful superconductors, the formulas of the NRSF and static conductivity derivative are obtained as

$$\sigma_{\text{NRSF}}^{\alpha;\beta\gamma} = \frac{1}{2\omega_1\omega_2} f^{\alpha\beta\gamma}, \quad (2.2.26)$$

$$\sigma_{\text{sCD}}^{\alpha;\beta\gamma} = -\frac{i}{4} \left(\frac{1}{\omega_1} \epsilon_{\alpha\gamma\delta} B_d^{\beta\delta} + \frac{1}{\omega_2} \epsilon_{\alpha\beta\delta} B_d^{\gamma\delta} \right). \quad (2.2.27)$$

In Eqs. (2.2.26) and (2.2.27), the NRSF $\hat{f}$ and Berry curvature derivative $\hat{B}_d$ are defined in terms of Green function [16].

2.2.2 Joint density of states

Before showing the nonlinear optical responses, we discuss the joint density of states (JDOS) in the Rashba superconductor model and analytically calculate it. The JDOS

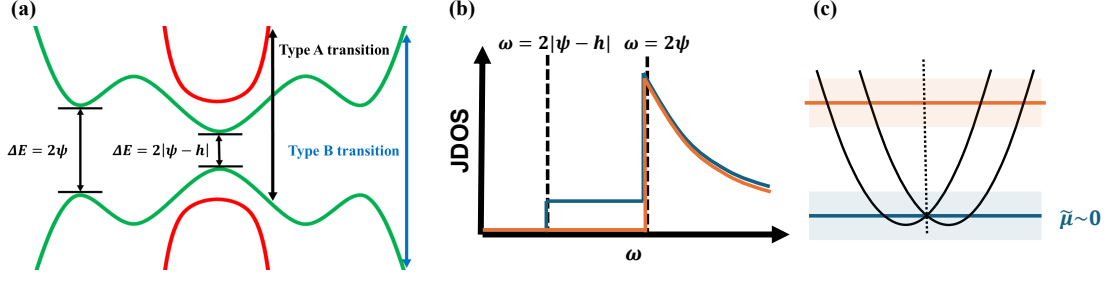


Figure 2.2: (a) Schematic illustration of the band structure and the optical transitions in the superconducting state. Energy bands of Bogoliubov quasiparticles. The type A and type B transitions give resonant contributions to the second-order optical responses. (b) Schematic illustration of the JDOS, which is derived from the type B transition. The color of the lines corresponds to the Fermi level illustrated in (c). A peak appears in the JDOS at $\omega = 2\psi$. In addition, a plateau (blue line) may appear between $\omega = 2|\psi - \hbar|$ and $\omega = 2\psi$, when the Fermi level is close to the Dirac point at the time-reversal invariant momentum (Γ point in this paper). (c) Approximate parabolic bands around the Γ point in the normal state. The nonlinear optical responses show characteristic behaviors under the magnetic field when the chemical potential measured from the Dirac point $\tilde{\mu}$ is small.

is an essential quantity for the nonlinear optical responses because the formulas of the normal photocurrent conductivity, Eqs. (2.2.17)-(2.2.20), contain the factor $\delta(\omega - E_{ba})$, which corresponds to the optical transition between the a and b bands. Below we show the characteristic frequency dependence of the JDOS. In addition, we identify the essential optical transition and approximately decompose the corresponding JDOS to $\tilde{J}_{B2(i)}$ and $\tilde{J}_{B2(ii)}$. These approximated JDOS show characteristic dependence on the magnetic field and chemical potential. In the later sections, the photocurrent conductivity is discussed based on the analysis of the JDOS.

We will show peculiar behaviors of the photocurrent conductivity around the topological transition. The condition for the topological superconductivity is Eq. (2.1.12). Thus, we mainly discuss the Rashba system with the Fermi level near the Dirac point, which is feasible for topological superconductivity.

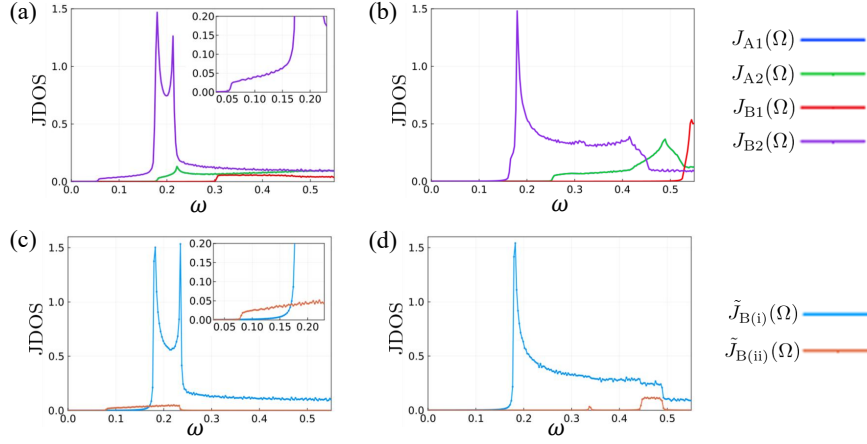


Figure 2.3: (Upper panels) The JDOS for the chemical potential (a) $\mu = -3.2$ and (b) -3.0 . The JDOS $J_{A1}(\omega)$, $J_{A2}(\omega)$, $J_{B1}(\omega)$, and $J_{B2}(\omega)$ correspond to the optical transitions $E_{3\mathbf{k}} \leftrightarrow E_{1\mathbf{k}}$, $E_{4\mathbf{k}} \leftrightarrow E_{2\mathbf{k}}$, $E_{4\mathbf{k}} \leftrightarrow E_{1\mathbf{k}}$, and $E_{3\mathbf{k}} \leftrightarrow E_{2\mathbf{k}}$, respectively. We set the parameters $t_1 = 1$, $t_2 = 0.2$, and $h = 0.05$. (Lower panels) Blue and orange lines plot the approximate JDOS, $\tilde{J}_{B2(i)}(\omega)$ and $\tilde{J}_{B2(ii)}(\omega)$, respectively. The effective chemical potential in (c) $\tilde{\mu} = 0$ and (d) 0.2 correspond to the panels (a) and (b), respectively. We adopt $t = 0.6$ consistent with the tight-binding parameters $t_1 = 1$ and $t_2 = 0.2$. In the cases (a) $\mu = -3.2$ and (c) $\tilde{\mu} = 0$, the Dirac point lies on the Fermi level. The characteristic frequencies are given as below. (d) $\omega = 2 \left| \sqrt{\xi_{\mathbf{k}=\mathbf{0}}^2 + \psi^2} - h \right| = 0.08$ and (e) 0.141 . (a) $\omega = 2 \left| \sqrt{\xi_{\mathbf{k}=\mathbf{0}}^2 + \psi^2} - h_{\text{eff}} \right| = 0.056$ and (b) 0.119 . In all the panels (a)-(f), $\omega = 2\psi = 0.18$. Reproduced from Fig. 3 of Ref. [57]. © 2024 American Physical Society

Numerical results of JDOS

We defined the JDOS, J_{A1} , J_{A2} , J_{B1} , and J_{B2} , as

$$J_{A1}(\omega) = \sum_{\mathbf{k}} \delta(\omega - E_{3\mathbf{k}} + E_{1\mathbf{k}}), \quad (2.2.28)$$

$$J_{A2}(\omega) = \sum_{\mathbf{k}} \delta(\omega - E_{4\mathbf{k}} + E_{2\mathbf{k}}), \quad (2.2.29)$$

$$J_{B1}(\omega) = \sum_{\mathbf{k}} \delta(\omega - E_{4\mathbf{k}} + E_{1\mathbf{k}}), \quad (2.2.30)$$

$$J_{B2}(\omega) = \sum_{\mathbf{k}} \delta(\omega - E_{3\mathbf{k}} + E_{2\mathbf{k}}), \quad (2.2.31)$$

where we define the eigenenergy of the quasiparticles $E_i(\mathbf{k})$ as $E_1(\mathbf{k}) \leq E_2(\mathbf{k}) \leq 0 \leq E_3(\mathbf{k}) \leq E_4(\mathbf{k})$. We use the notation $A_{\mathbf{k}} \equiv A(\mathbf{k})$ for simplicity. The JDOS with subscript B, namely, J_{B1} and J_{B2} , correspond to the type B transition specified in Fig. 2.2(a). Figure 2.3(a) and 2.3(b) show the JDOS for $\mu = -3.2$ and -3.0 , respectively. We see that the JDOS $J_{B2}(\omega)$ is dominant at low energies. Thus, it is expected that the type B optical transition between the $E_{2\mathbf{k}}$ and $E_{3\mathbf{k}}$ bands dominates the superconducting nonlinear responses to the irradiation of low-frequency light.

In our model, the Dirac point exists at $\mathbf{k} \simeq \mathbf{0}$. When the magnetic field is absent, the chemical potential $\mu = -3.2$ lies on the Dirac point [blue lines in Fig. 2.2(c)], while $\mu = -3.0$ lie above the Dirac point [a orange line in Fig. 2.2(c)]. The chemical potential dependence of the JDOS $J_{B2}(\omega)$ was illustrated in Fig. 2.2(b) based on the results in Figs. 2.3(a) and 2.3(b). Specifically, the JDOS has characteristic structures. First, a sharp peak exists around $\omega \simeq 2\psi = 0.18$ independent of the chemical potential. Second, Fig. 2.2(a) ($\mu = -3.2$) show finite $J_{B2}(\omega)$ below the frequency $\omega = 2\psi$, while Fig. 2.2 (b) does not show it. The origin of the similarities and difference between JDOS spectra for different chemical potentials is explained below.

Approximate JDOS

To analyze the system with a Fermi level near the Dirac point it is essential to understand the JDOS illustrated in Fig. 2.2(b) and obtained in Fig. 2.3. For this purpose, we introduce the approximate JDOS, $\tilde{J}_{B2(i)}(\omega)$ and $\tilde{J}_{B2(ii)}(\omega)$, into which $J_{B2}(\omega)$ is approximately decomposed. Here, we approximate the normal state Hamiltonian with a focus on the Γ point ($\mathbf{k} = \mathbf{0}$) of the Brillouin zone as

$$\xi_{\mathbf{k}} = t(k_x^2 + k_y^2) - \tilde{\mu}, \quad (2.2.32)$$

$$\mathbf{g}_{\mathbf{k}} = \alpha(k_y, -k_x, 0), \quad (2.2.33)$$

$$\tilde{\mu} \equiv \mu + 4(t_1 + t_2), \quad (2.2.34)$$

where $\tilde{\mu}$ is the effective chemical potential indicating the difference of the Fermi level from the Dirac point. For our choice of parameters, $\mu = -3.2$ corresponds to $\tilde{\mu} = 0$ while $\mu = -3.0$ to $\tilde{\mu} = 0.2$. For simplicity, we ignore the total momentum of Cooper pairs and set $2\mathbf{q} = \mathbf{0}$ in the discussion of the approximate JDOS.

First, we derive the approximate JDOS $\tilde{J}_{\text{B2(i)}}(\omega)$, which gives a sharp peak of $J_{\text{B2}}(\omega)$ around $\omega = 2\psi$. In the derivation, we ignore $\mathbf{g}_{\mathbf{k}} \cdot \mathbf{h}$ because its effect is almost canceled between E_2 and E_3 in the energy difference ΔE_{32} . Then we obtain the energies of Bogoliubov quasiparticles as

$$E_{\mathbf{k}} = \pm \sqrt{\xi_{\mathbf{k}}^2 + g_{\mathbf{k}}^2 + h^2 + \psi^2 \pm 2\sqrt{\xi_{\mathbf{k}}^2(g_{\mathbf{k}}^2 + h^2) + \psi^2 h^2}}. \quad (2.2.35)$$

The approximate energy satisfies the symmetry $E_{\mathbf{k}} = E_{-\mathbf{k}}$ because we ignore $\mathbf{g}_{\mathbf{k}} \cdot \mathbf{h}$. When $|\xi_{\mathbf{k}}||\mathbf{g}_{\mathbf{k}}| \gg \psi h$, the energies of Bogoliubov quasiparticles are furthermore approximated by

$$E_{\mathbf{k}} = \pm \sqrt{\left(|\xi_{\mathbf{k}}| \pm \sqrt{g_{\mathbf{k}}^2 + h^2}\right)^2 + \psi^2}. \quad (2.2.36)$$

To estimate the JDOS, we consider an electron band of $E_{3\mathbf{k}}$,

$$E_{3\mathbf{k}} = \sqrt{\left(|\xi_{\mathbf{k}}| - \sqrt{g_{\mathbf{k}}^2 + h^2}\right)^2 + \psi^2}. \quad (2.2.37)$$

Since $E_{2\mathbf{k}}$ and $E_{3\mathbf{k}}$ are related to each other under the charge-conjugation transformation $E_{2\mathbf{k}} = -E_{3-\mathbf{k}}$, the energy difference of the transition between the electron and hole bands is given by

$$\Delta E_{\mathbf{k}} = E_{3\mathbf{k}} - E_{2\mathbf{k}} = 2\sqrt{\left(|\xi_{\mathbf{k}}| - \sqrt{g_{\mathbf{k}}^2 + h^2}\right)^2 + \psi^2}. \quad (2.2.38)$$

The approximate JDOS $\tilde{J}_{\text{B2(i)}}(\omega)$ is introduced corresponding to Eq. (2.2.38),

$$\tilde{J}_{\text{B2(i)}}(\omega) = \sum_{\mathbf{k}}' (\omega - \Delta E_{\mathbf{k}}), \quad (2.2.39)$$

where the integration is carried out on the region $|\xi_{\mathbf{k}}||\mathbf{g}_{\mathbf{k}}| > \psi h$. When we assume $g_{\mathbf{k}} \gg h$, which is satisfied for large k , $\tilde{J}_{\text{B2(i)}}(\omega)$ shows the divergence at $\omega = 2\psi$. In fact, $\tilde{J}_{\text{B2(i)}}(\omega)$ reproduces the sharp peak of the JDOS around $\omega = 2\psi$, as shown in Figs. 2.3(c) and 2.3(d). We also see the peaks at $\Omega \simeq 0.22$ in Figs. 2.3(a) and 2.3(c). These peak positions are sensitive to the spin-orbit coupling parameter α .

Next, we introduce the approximate JDOS $\tilde{J}_{\text{B2(ii)}}(\omega)$ base on another assumption $\psi h \gg |\xi_{\mathbf{k}}||\mathbf{g}_{\mathbf{k}}|$. In this case, we obtain the electron band of $E_{3\mathbf{k}}$,

$$E_{3\mathbf{k}} = \sqrt{\xi_{\mathbf{k}}^2 + g_{\mathbf{k}}^2 + h^2 + \psi^2 - 2h\sqrt{\xi_{\mathbf{k}}^2 + \psi^2}}. \quad (2.2.40)$$

Then, the energy difference for the optical transition between the electron and hole bands is given by

$$\Delta E_{\mathbf{k}} = 2\sqrt{\xi_{\mathbf{k}}^2 + g_{\mathbf{k}}^2 + h^2 + \psi^2 - 2h\sqrt{\xi_{\mathbf{k}}^2 + \psi^2}}. \quad (2.2.41)$$

Thus, the approximate JDOS $\tilde{J}_{\text{B2(ii)}}(\omega)$ is introduced as

$$\tilde{J}_{\text{B2(ii)}}(\omega) = \sum_{\mathbf{k}}'' \delta \left(\omega - 2\sqrt{\xi_{\mathbf{k}}^2 + g_{\mathbf{k}}^2 + h^2 + \psi^2 - 2h\sqrt{\xi_{\mathbf{k}}^2 + \psi^2}} \right). \quad (2.2.42)$$

where the integration is carried out on the region $\psi h > |\xi_{\mathbf{k}}||g_{\mathbf{k}}|$. By the integration around $\mathbf{k} = \mathbf{0}$, $\tilde{J}_{\text{B2(ii)}}(\omega)$ is approximated by

$$\tilde{J}_{\text{B2(ii)}}(\omega) = \begin{cases} 0 & \left(0 \leq \omega < 2\left|\sqrt{\tilde{\mu}^2 + \psi^2} - h\right| \right) \\ \frac{\Omega}{8\pi\tilde{\alpha}^2} & \left(2\left|\sqrt{\tilde{\mu}^2 + \psi^2} - h\right| \leq \omega \right) \end{cases}, \quad (2.2.43)$$

with

$$\tilde{\alpha} \equiv \sqrt{\alpha^2 - 2t\tilde{\mu} + \frac{2t\tilde{\mu}h}{\sqrt{\tilde{\mu}^2 + \psi^2}}}. \quad (2.2.44)$$

We see that the JDOS $\tilde{J}_{\text{B2(ii)}}(\omega)$ is finite above $\omega = 2\left|\sqrt{\tilde{\mu}^2 + \psi^2} - h\right|$. We expect that the contribution of $\tilde{J}_{\text{B2(ii)}}(\omega)$ is important in the low-frequency region because it can be finite for the frequency ω smaller than 2ψ contrary to $\tilde{J}_{\text{B2(i)}}(\omega)$ when $|\tilde{\mu}|$ is sufficiently small. Corresponding to this fact, we will see the nonlinear optical responses in the region $2|\psi - h| \leq \omega \leq 2\psi$ when the Fermi level lies on the Dirac point. As shown in Fig. 2.3(c), $\tilde{J}_{\text{B2(ii)}}(\omega)$ for $\tilde{\mu} = 0$ shows a plateau starting from $\omega = 2|\psi - h|$. A similar structure is seen in the numerical results of the JDOS [Fig. 2.3(a)]. Indeed, the JDOS $J_{\text{B2}}(\omega)$ can be approximated by summation of $\tilde{J}_{\text{B2(i)}}(\omega)$ and $\tilde{J}_{\text{B2(ii)}}(\omega)$, as $J_{\text{B2}}(\omega) \simeq \tilde{J}_{\text{B2(i)}}(\omega) + \tilde{J}_{\text{B2(ii)}}(\omega)$. This feature was illustrated in Fig. 2.2(b).

Comparison of JDOS and approximate JDOS

Here, we compare the JDOS with the approximate JDOS. First, let us compare Fig. 2.3(a) with Fig. 2.3(c), where the chemical potential lies on the Dirac point. The plateau of $J_{\text{B2}}(\omega)$ as $\omega < 2\psi$ is attributed to $\tilde{J}_{\text{B2(ii)}}(\omega)$, and the sharp peak of $J_{\text{B2}}(\omega)$ at $\omega = 2\psi$ is attributed to $\tilde{J}_{\text{B2(i)}}(\omega)$. Thus, we see a qualitative agreement between the numerically calculated JDOS and the approximate JDOS.

Contrary to the above case, we do not see the contribution of $\tilde{J}_{\text{B2(ii)}}(\omega)$ in the JDOS for $\mu = -3.0$ [Fig. 2.2(c)] corresponding to an effective chemical potential $\tilde{\mu} = 0.2$ deviating from the Dirac point. The sharp peak of the JDOS still appears around $\omega = 2\psi$ and is

attributed to $\tilde{J}_{B2(i)}(\omega)$. On the other hand, $\tilde{J}_{B2(ii)}(\omega)$ vanishes in the low-frequency region $\omega \leq 2\psi$ because a finite contribution is obtained only above $\omega = 2|\sqrt{\tilde{\mu}^2 + \psi^2} - h| \simeq 0.34 > 2\psi$.

Based on the above results, we expect characteristic behaviors in the nonlinear optical responses, such as the peculiar magnetic field dependence, when the Fermi level is close to the Dirac point. As indicated by Eq. (2.2.43), finite photocurrent conductivity appears in the in-gap region $\omega < 2\psi$ under the magnetic fields. On the other hand, a sharp peak of the JDOS around $\omega = 2\psi$ is not sensitive to the magnetic field except for the change of the pair potential ψ , and therefore, we expect that the nonlinear optical responses in this frequency range are robust under the magnetic field.

2.2.3 Numerical demonstration

In this subsection, we numerically demonstrate the nonlinear optical conductivity in the Rashba superconductors. In the two-dimensional superconductors with the C_{4v} crystal structure, our Rashba model, all the second-order optical response are prohibited at the zero magnetic field. However, the second-order responses may occur under the magnetic field, which breaks not only the $\mathcal{T}$ symmetry but also the crystalline symmetry. We will show the field-induced nonlinear optical responses in the following.

For a quantitative estimation, we set $t_1 = 1\text{eV}$ and calculate the response coefficients in the SI unit. Thus, the results of the nonlinear conductivity are given in the unit AV^{-2} . Numerical calculations are performed on the N^2 -discretized Brillouin zone ($N = 3000$). For numerical convergence, we replace the parameter η with a phenomenological scattering rate $\gamma = 2.0 \times 10^{-4}$ and introduce a finite temperature $T = 10^{-4}$ for the Fermi-Dirac distribution function.

Characteristic frequency dependence and constraints due to symmetry

We show the photocurrent generation and SHG under the in-plane magnetic field ($\theta = 0$) in Fig. 2.4. It should be noted that some components vanish due to the constraints discussed below. First, the constraint

$$\sigma_{\text{PC}}^{\alpha;\beta\gamma} = \left(\sigma_{\text{PC}}^{\alpha;\gamma\beta}\right)^*, \quad \sigma_{\text{SHG}}^{\alpha;\beta\gamma} = \sigma_{\text{SHG}}^{\alpha;\gamma\beta}, \quad (2.2.45)$$

has to be satisfied by definition. This constraint requires $\Im[\sigma_{\text{PC}}^{\alpha;\beta\beta}] = 0$. Second, the symmetry of the system prohibits some components of the nonlinear optical responses. In this respect, the magnetic field plays a key role. In our model, the C_{4v} symmetry of the crystal structure lowers under the magnetic field, and thus the magnetic field is indispensable because the C_{4v} point group symmetry prohibits the second-order nonlinear

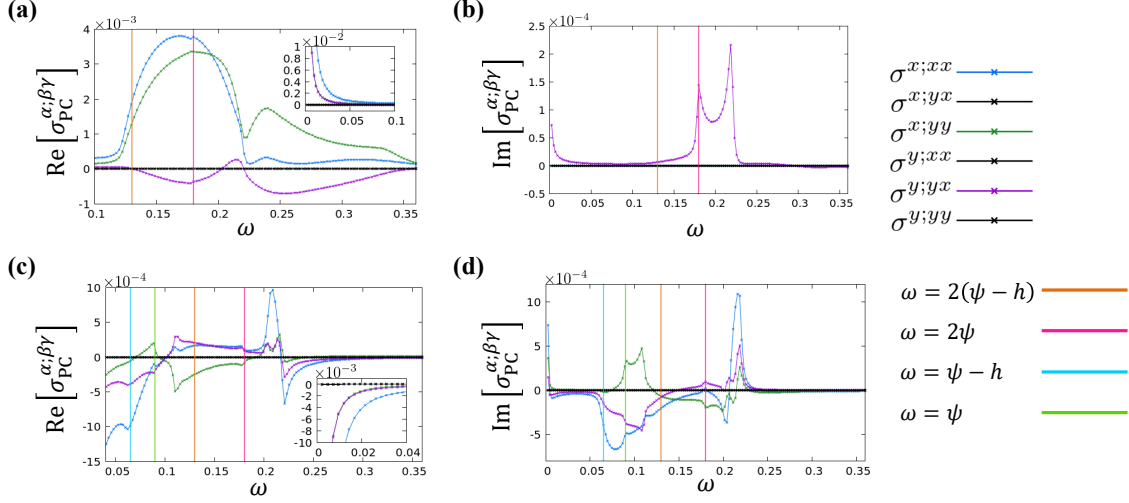


Figure 2.4: Frequency dependence of the photocurrent generation and the SHG under the in-plane magnetic field ($\theta = 0^\circ$). The unit of the vertical axis is (ÅV^{-2}) . (a) Real part $\text{Re}[\sigma_{\text{PC}}^{\alpha;\beta\gamma}]$ and (b) imaginary part $\text{Im}[\sigma_{\text{PC}}^{\alpha;\beta\gamma}]$ of the photocurrent generation. (c) Real part $\text{Re}[\sigma_{\text{SHG}}^{\alpha;\beta\gamma}]$ and (d) imaginary part $\text{Im}[\sigma_{\text{SHG}}^{\alpha;\beta\gamma}]$ of the SHG. We set the parameters $h = 0.025$, $\psi = 0.09$, and $\mu = -3.2$. Note that $\sigma^{x;yx} = \sigma^{y;xx} = \sigma^{y;yy} = 0$ since these components are prohibited by the $m'm2'$ magnetic point group symmetry. The vertical lines illustrate the frequencies $\Omega = 2\psi, 2|\psi - h|$ for the photocurrent generation and $\Omega = \psi, 2\psi, \psi - h, 2|\psi - h|$ for the SHG. Reproduced from Fig. 6 of Ref. [57]. © 2024 American Physical Society

Table 2.3: Constrains of second-order optical responses and the symmetry class with θ being the angle between the magnetic field and the two-dimensional plane [$\mathbf{h} = (0, h \cos \theta, h \sin \theta)$]. The subscript ‘ $\mathcal{T}$ -even’ means the contribution which is even under the $\mathcal{T}$ operation (see also Table 2.2).

Angle θ	Symmetry	Constraints
$\mathbf{h} \parallel \hat{\mathbf{y}} (\theta = 0^\circ)$	$m'm2'$	$\sigma^{x;yx} = \sigma^{y;xx} = \sigma^{y;yy} = 0$, $\sigma_{\mathcal{T}\text{-even}}^{\alpha;\beta\gamma} = 0$ for all α, β, γ
$0^\circ < \theta < 90^\circ$	m'	$\sigma_{\mathcal{T}\text{-even}}^{x;xx} = \sigma_{\mathcal{T}\text{-even}}^{x;yy} = \sigma_{\mathcal{T}\text{-even}}^{y;yx} = 0$
$\mathbf{h} \parallel \hat{\mathbf{z}} (\theta = 90^\circ)$	$4m'm'$	$\sigma^{\alpha;\beta\gamma} = 0$ for all α, β, γ

response in two-dimensional systems. The relation between the symmetry constraints and the magnetic fields is summarized in Table 2.3 and discussed in the following. First, when the magnetic field $\mathbf{h} = (0, h \cos \theta, h \sin \theta)$ is parallel to the y axis ($\theta = 0^\circ$), the model is characterized by the $m'm2'$ magnetic point group symmetry. The symmetry prohibits the nonlinear conductivities $\sigma^{x;yx}$, $\sigma^{y;xx}$, and $\sigma^{y;yy}$. Figure 2.4 is consistent with these constraints. Moreover, all the $\mathcal{T}$ -even contributions symmetric under the $\mathcal{T}$ operation (see Table 2.2 for classification of the photocurrent conductivity based on the mechanism and symmetry) disappear in the two-dimensional system due to the $\mathcal{TC}_{2z}$ symmetry. Second, when θ satisfies $0^\circ < \theta < 90^\circ$, the model has the $\mathcal{T}m_x$ symmetry, which is classified into the magnetic point group m' . The $\mathcal{T}$ -even contributions of $\sigma^{x;yx}$, $\sigma^{y;xx}$, and $\sigma^{y;yy}$ disappear due to the symmetry. Finally, when the magnetic field is perpendicular to the xy plane ($\theta = 90^\circ$), the symmetry of the system is $4m'm'$, and all the components of nonlinear conductivity vanish in the two-dimensional system due to the C_{2z} rotation symmetry.

In Fig. 2.4, we see a characteristic frequency dependence of superconducting nonlinear responses. First, in the low-frequency region, the divergent nonlinear responses are observed in the real part of $\sigma_{PC}^{\alpha;\beta\gamma}$ and $\sigma_{SHG}^{\alpha;\beta\gamma}$. The NRSF term $\sigma_{NRSF}^{\alpha;\beta\gamma}$, which is odd under the $\mathcal{T}$ symmetry, causes the anomalous divergent behaviors. The static conductivity derivative term $\sigma_{sCD}^{\alpha;\beta\gamma}$ disappears because this term is even for the $\mathcal{T}$ symmetry. Although the imaginary part also shows a weakly diverging behavior at low frequencies, this behavior is artificial and comes from the phenomenological treatment of the scattering rate [16].

Second, the type B transition illustrated in Fig. 2.4(a) gives the characteristic resonant contribution to the photocurrent conductivity and SHG. This contribution is often characterized by the superconducting gap. The peak around $\omega = 2\psi$ is a typical behavior and observed in Figs. 2.4(a) and 2.4(b). Contrary to the photocurrent, not only the transition $\Delta E \sim \omega$ but also $\Delta E \sim 2\omega$ may give rise to essential contributions to the second harmonic generation, where ΔE is an energy difference between transition bands. Figures 2.4(c) and 2.4(d) show the peak structure and sign reversal at $\omega = \psi$. The frequency dependence of the SHG is more complex than that of the photocurrent generation. In the remaining part, we discuss the photocurrent conductivity in detail.

Unique behaviors due to Dirac point

We will show the characteristic behavior of the nonlinear optical responses in Rashba systems with a chemical potential near the Dirac point. For this purpose, we focus on the resonant components of the photocurrent, which the JDOS characterizes. We investigate in detail the dependence on the magnetic field. Consistent with our conjecture in Sec. 2.2.2, the presence of the Dirac point near the Fermi level gives rise to unique behaviors in the nonlinear responses in the superconducting states. In the following, we verify the

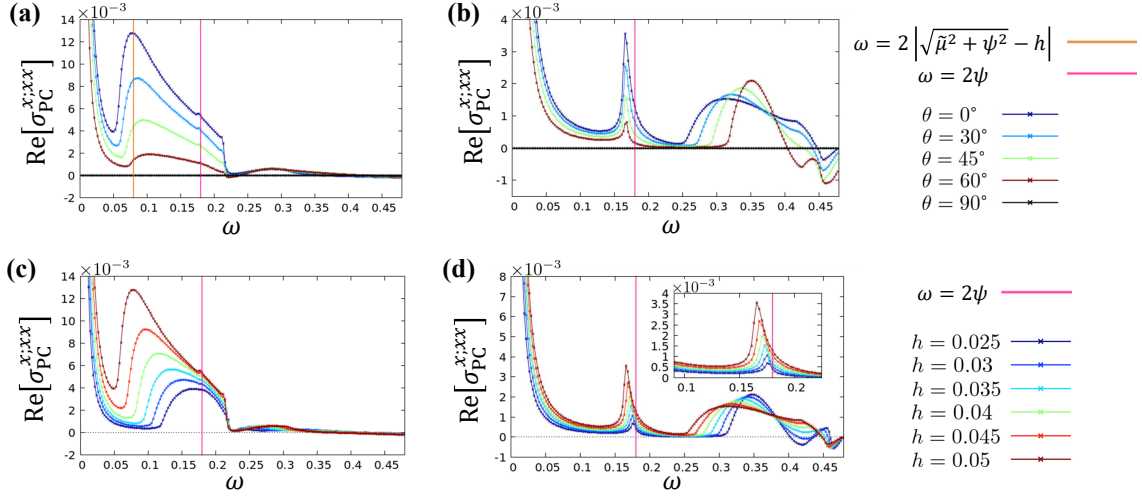


Figure 2.5: Photocurrent conductivity $\text{Re}[\sigma_{\text{PC}}^{xx}]$ with varying the magnetic field $\mathbf{h}$. The unit of the vertical axis is (ÅV^{-2}) . (a), (b) The angle of the magnetic field is varied by fixing the magnitude $h = 0.05$. (c), (d) The magnitude of the magnetic field is varied by fixing the angle $\theta = 0^\circ$. The chemical potential is set as (a), (c) $\mu = -3.2$ while (b), (d) $\mu = -3.0$. Reproduced from Figs. 8 and 9 of Ref. [57]. © 2024 American Physical Society

conjecture with numerical calculations.

First, we discuss the angle θ of the magnetic field $\mathbf{h} = (0, h \cos \theta, h \sin \theta)$. Figure 2.5(a) plots the real part of the photocurrent conductivity σ_{PC}^{xx} for various angles. Here, we see a consequence of the symmetry constraint. The photocurrent conductivity disappears under the perpendicular magnetic field with $\theta = 90^\circ$ because the C_{2z} rotation symmetry prohibits the second-order optical responses. Therefore, the magnitude of the photocurrent conductivity becomes smaller when the angle approaches $\theta = 90^\circ$. For angles different from $\theta = 90^\circ$, we see qualitatively the same frequency dependence. In Fig. 2.5(a) with $\mu = -3.2$, the photocurrent conductivity is enhanced between $\omega = 2|\psi - h|$ ($= 2|\sqrt{\tilde{\mu}^2 + \psi^2} - h|$) and $\omega = 2\psi$.

Next, we show the change in the photocurrent conductivity when varying the magnitude of the magnetic field. Figure 2.5(c) plot the real part of the photocurrent conductivity σ_{PC}^{xx} with varying the magnitude. For $\mu = -3.2$ on the Dirac point [Fig. 2.5(c)], the photocurrent conductivity begins rising at a lower frequency with a larger magnetic field. This is consistent with the JDOS, which is finite above $\omega \simeq 2|\psi - h|$ and attributed to the contribution of $\tilde{J}_{\text{B2(ii)}}(\omega)$.

However, the above peculiar behaviors of the photocurrent conductivity disappear when the chemical potential is far away from the Dirac point. This is expected from the analysis of the JDOS. Because the unique contribution in the in-gap region $\omega < 2\psi$ disappears at

$\mu = -3.0$, we expect that the contribution of $\tilde{J}_{B2(ii)}(\omega)$ is dominant. In Figs. 2.5(b) and 2.5(d), where we plot $\text{Re}[\sigma_{PC}^{xx}]$ for $\mu = -3.0$, the photocurrent conductivity shows a peak around $\omega = 2\psi$, and the position of the peak does not significantly change with varying the angle and magnitude of the magnetic field. These results are consistent with the above expectation that the photocurrent conductivity is mainly related to the JDOS $\tilde{J}_{B2(i)}(\omega)$ when the chemical potential is not close to the Dirac point.

We summarize the results in this subsection. For chemical potentials near the Dirac point, a component of the JDOS from the region $\mathbf{k} \simeq \mathbf{0}$, namely $\tilde{J}_{B2(ii)}(\omega)$, gives significant contributions to the photocurrent conductivity in the superconducting state. Since these contributions are sensitive to magnetic fields, the photocurrent conductivity shows characteristic behavior in the in-gap region $\omega < 2\psi$. On the other hand, the contribution of $\tilde{J}_{B2(i)}(\omega)$ is dominant when the chemical potential is not in the vicinity of the Dirac point. This contribution shows a sharp peak around $\omega = 2\psi$, which is robust to changes in the magnetic field. The decomposition of the JDOS to the approximate JDOS $\tilde{J}_{B2(i)}(\omega)$ and $\tilde{J}_{B2(ii)}(\omega)$ is helpful to elucidate the photocurrent conductivity in noncentrosymmetric superconductors in the in-gap frequency range $\omega \lesssim 2\psi$.

2.3 Nonlinear optical conductivity around topological transition

Next, we investigate the relation between superconducting nonlinear responses and topological superconductivity. We show that the photocurrent conductivity and the SHG coefficient dramatically change when the Rashba superconductor becomes the topological phase.

We have considered s -wave Rashba superconductors with the Zeeman field. The Rashba superconductor becomes a topological superconducting state when the Zeeman field h is larger than the critical value h_c . When the magnetic field is perpendicular to the plane, the critical value h_c is obtained as $h^c = \sqrt{\psi^2 + \xi(0,0)^2} = \sqrt{\tilde{\mu}^2 + \psi^2}$ from the condition (2.1.12).

First, we investigate the photocurrent conductivity around the topological transition between the topologically trivial and nontrivial superconducting phases. In the previous papers [68, 71, 72], it was shown that the topological transition occurs under the magnetic field perpendicular to the plane. In the following, we consider tilted magnetic fields to the plane because the photocurrent conductivity vanishes under the perpendicular magnetic field. We set the angle θ as $\theta = 60^\circ$. Because the topological properties are robust against small changes in the Hamiltonian, the topological transition occurs even though the tilted magnetic fields. Gap closing is one of the characterizations of the topological transition.

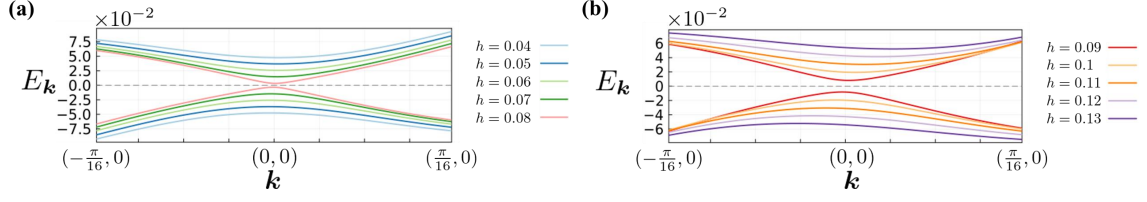


Figure 2.6: The energy band of Bogoliubov quasiparticles around the Γ point [$\mathbf{k} = \mathbf{0}$] with varying the magnitude of the magnetic field. We set $\mu = -3.2$, $\psi = 0.09$, and $\theta = 60^\circ$. From Fig. 12 of Ref. [57]. © 2024 American Physical Society

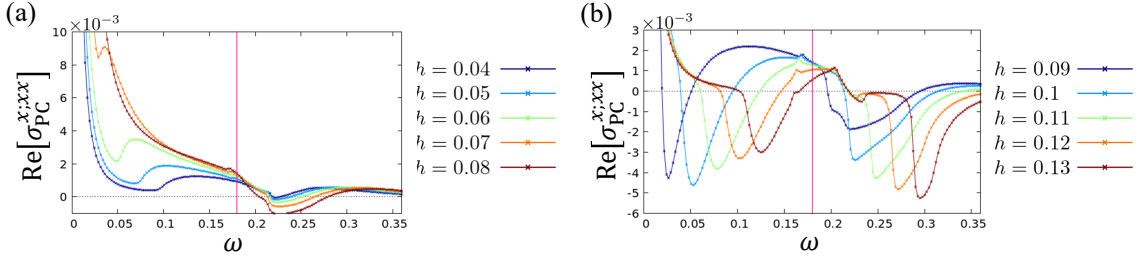


Figure 2.7: The photocurrent conductivity for various magnitudes of the magnetic field. The unit of the vertical axis is (ÅV^{-2}) . We show $\text{Re}[\sigma_{\text{PC}}^{x;xx}]$ for (a) $0.04 \leq h \leq 0.08$ and (b) $0.09 \leq h \leq 0.13$. Reproduced from Fig. 13 of Ref. [57]. © 2024 American Physical Society

Figure 2.6 shows the energy band around $\mathbf{k} = \mathbf{0}$ when we set $\mu = -3.2$ and $\psi = 0.09$. As we enlarge the magnitude of the magnetic field in $0.04 \leq h \leq 0.08$, the energy gap becomes smaller [Fig. 2.6(a)]. On the other hand, the energy gap opens as the magnetic field is further increased in $0.09 \leq h \leq 0.13$ [Fig. 2.6(b)]. These results indicate that the gap closing and topological transition occur in the region $0.08 \leq h \leq 0.09$.

Figure 2.7 shows the photocurrent conductivity $\sigma_{\text{PC}}^{x;xx}$ for the magnetic fields (a) $0.04 \leq h \leq 0.08$ and (b) $0.09 \leq h \leq 0.13$. We focus on the resonant components in the low-frequency region $\omega < 2\psi$. In the topologically trivial region $0.04 \leq h \leq 0.08$, the photocurrent conductivity begins to increase at a lower frequency with a larger magnetic field [Fig. 2.7(a)]. When the magnetic field is further increased in the topological superconducting phase, the photocurrent conductivity is negatively enhanced and shows peaks at higher frequencies with larger magnetic fields [Fig. 2.7(b)]. The drastic changes in frequency dependence and sign are associated with the closing of the energy gap around $\mathbf{k} = \mathbf{0}$ (Fig. 2.6). It is naturally expected that these low-frequency contributions originate from the JDOS $\tilde{J}_{\text{B2(ii)}}$. We would like to emphasize the sign change of the resonant photocurrent before and after the topological transition. Below, we discuss the impact of the topological transition and attribute the sign change to band inversion, which plays a key role in various

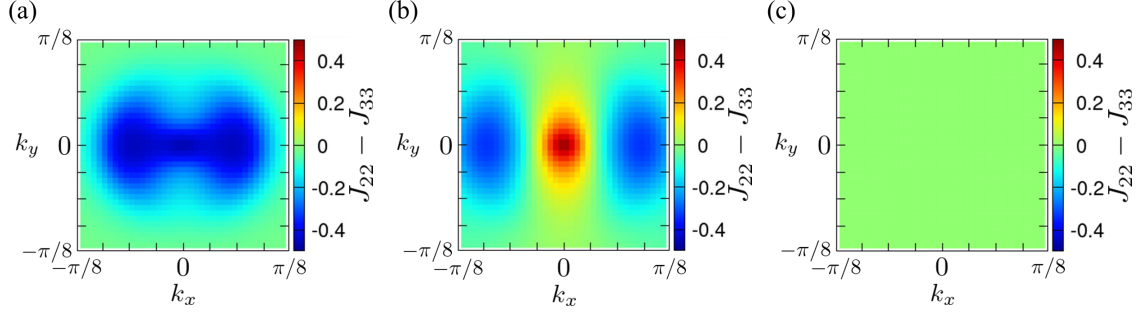


Figure 2.8: Distribution of $J_{22}^x - J_{33}^x$ in the $\mathbf{k}$ space. (a), (b) We fix the angle of the magnetic field $\theta = 60^\circ$ and set the magnitude (a) $h = 0.06$ and (b) $h = 0.1$ below and above the topological transition, respectively. (c) We set $\theta = 90^\circ$ and $h = 0.05$. We see that $J_{22}^x - J_{33}^x$ vanishes. From Fig. 14 of Ref. [57]. © 2024 American Physical Society

topological transitions.

In the low-frequency regime, the magnetic injection current originating from the B2 transition $\sigma_{\text{Minj(B2)}}^{x;xx}$ gives dominant contributions to the photocurrent conductivity $\sigma_{\text{PC}}^{x;xx}$. In the zero temperature limit, $\sigma_{\text{Minj(B2)}}^{x;xx}$ is given by

$$\sigma_{\text{Minj(B2)}}^{x;xx} = -\frac{\pi}{4\eta} \sum_{\mathbf{k}} (J_{22}^x - J_{33}^x) g_{32}^{xx} \delta(\omega - E_{32}). \quad (2.3.1)$$

The quantum metric g_{32}^{xx} is finite and positive. Thus, the sign of $\sigma_{\text{Minj(B2)}}^{x;xx}$ is determined by the difference of the paramagnetic current $J_{22}^x - J_{33}^x$. The sign of $J_{22}^x - J_{33}^x$ around $\mathbf{k} = \mathbf{0}$ is opposite between the trivial state [Fig. 2.8(a)] and the topological state [Fig. 2.6(b)]. This behavior causes the sign change of the photocurrent conductivity $\sigma_{\text{PC}}^{x;xx}$.

The sign change of $J_{22}^x - J_{33}^x$ is caused by the band inversion. At the Γ point ($\mathbf{k} = \mathbf{0}$), the eigenenergies of Bogoliubov quasiparticles $E_{2\mathbf{k}}$ and $E_{3\mathbf{k}}$ are obtained as

$$(E_{2\mathbf{k}}, E_{3\mathbf{k}}) = \begin{cases} (E^{(-)}, E^{(+)}) & (\sqrt{\tilde{\mu}^2 + \psi^2} > h) \\ (E^{(+)}, E^{(-)}) & (\sqrt{\tilde{\mu}^2 + \psi^2} < h) \end{cases}, \quad (2.3.2)$$

where we define $E^{(-)}$ and $E^{(+)}$ as

$$E^{(-)} = -\sqrt{\xi_{\mathbf{k}=\mathbf{0}}^2 + \psi^2} + h, \quad E^{(+)} = \sqrt{\xi_{\mathbf{k}=\mathbf{0}}^2 + \psi^2} - h. \quad (2.3.3)$$

This indicates the band inversion between the $E^{(-)}$ band and the $E^{(+)}$ band at the topological transition. Note that the subscript a of $E_{a\mathbf{k}}$ is defined as $E_{1\mathbf{k}} \leq E_{2\mathbf{k}} \leq 0 \leq E_{3\mathbf{k}} \leq E_{4\mathbf{k}}$. For the $E^{(-)}$ and $E^{(+)}$ bands, the J_{aa}^α is obtained as

$$J_{aa}^\alpha = \partial_\alpha \xi_{\mathbf{k}} + \partial_\alpha \mathbf{g}_{\mathbf{k}} \cdot \hat{\mathbf{h}} \quad (E^{(-)} \text{ band}), \quad (2.3.4)$$

$$J_{aa}^\alpha = \partial_\alpha \xi_{\mathbf{k}} - \partial_\alpha \mathbf{g}_{\mathbf{k}} \cdot \hat{\mathbf{h}} \quad (E^{(+)} \text{ band}). \quad (2.3.5)$$

Thus, $J_{22}^x - J_{33}^x$ is given by

$$J_{22}^x - J_{33}^x = \begin{cases} 2(\partial_x \mathbf{g}_\mathbf{k} \cdot \hat{\mathbf{h}}) & (\sqrt{\tilde{\mu}^2 + \psi^2} > h) \\ -2(\partial_x \mathbf{g}_\mathbf{k} \cdot \hat{\mathbf{h}}) & (\sqrt{\tilde{\mu}^2 + \psi^2} < h) \end{cases}. \quad (2.3.6)$$

Because the $E^{(-)}$ and $E^{(+)}$ bands are inverted at the topological transition, the sign of $J_{22}^x - J_{33}^x$ changes resulting in the sign change of the photocurrent conductivity.

In the normal state, the diagonal element of the paramagnetic current operator J_{aa}^α is equivalent to the group velocity. With the Hellmann-Feynman theorem, the group velocity v_a^α is given by $v_a^\alpha = \partial_\alpha E_a = J_{aa}^\alpha$. In the superconducting state, the Hellmann-Feynman relation fails in the Bogoliubov de-Gennes formalism. However, the J_{aa}^α is closely related to the group velocity of electron and hole bands in the normal state. Below, we discuss the origin of $J_{22}^x - J_{33}^x$ from the perspective of the normal state band structure. Around $\mathbf{k} = \mathbf{0}$, the J_{aa}^α in the superconducting state is written as

$$J_{aa}^\alpha = w_- v_{e+}^\alpha - w_+ v_{h-}^\alpha \quad (E^{(-)} \text{ band}), \quad (2.3.7)$$

$$J_{aa}^\alpha = w_+ v_{e-}^\alpha - w_- v_{h+}^\alpha \quad (E^{(+)} \text{ band}), \quad (2.3.8)$$

where we define $w_\pm$ as

$$w_- = \frac{-\xi_\mathbf{k} + u}{2u}, \quad w_+ = \frac{\xi_\mathbf{k} + u}{2u}, \quad (2.3.9)$$

with $u = \sqrt{\xi_\mathbf{k}^2 + \psi^2}$. We introduce $v_{e\pm}^\alpha$ and $v_{h\pm}^\alpha$ as the group velocity of the electron band $\epsilon_\mathbf{k}^{e\pm} = \xi_\mathbf{k} \pm |\mathbf{g}_\mathbf{k} + \mathbf{h}|$ and the hole band $\epsilon_\mathbf{k}^{h\pm} = -\epsilon_{-\mathbf{k}}^{e\pm}$ in the normal state, respectively. In the normal state ($\psi = 0$), Eqs. (2.3.7) and (2.3.8) reproduce the Hellmann-Feynman relation. The difference $|J_{22}^\alpha - J_{33}^\alpha|$ is rewritten with $v_{e\pm}^\alpha$ and $v_{h\pm}^\alpha$ as

$$|J_{22}^\alpha - J_{33}^\alpha| = |w_- (v_{e+}^\alpha + v_{h+}^\alpha) - w_+ (v_{e-}^\alpha + v_{h-}^\alpha)|. \quad (2.3.10)$$

The difference in the effective currents between the electron and hole bands, $ev_{e\pm}^\alpha \neq -ev_{h\pm}^\alpha$, is essential for finite $|J_{22}^\alpha - J_{33}^\alpha|$. Note that we write the charge $e = 1$ explicitly here.

From the above discussion, we notice that the nonreciprocity in the band structure, namely $\epsilon_\mathbf{k}^{e\pm} \neq \epsilon_{-\mathbf{k}}^{e\pm}$, is essential for the difference $|J_{22}^\alpha - J_{33}^\alpha|$. Indeed, when we assume $\epsilon_\mathbf{k}^{e\pm} = \epsilon_{-\mathbf{k}}^{e\pm}$, the group velocity of the hole bands is obtained as

$$v_{h\pm}^\alpha = \partial_\alpha (-\epsilon_{-\mathbf{k}}^{e\pm}) = -\partial_\alpha \epsilon_\mathbf{k}^{e\pm} = -v_{e\pm}^\alpha, \quad (2.3.11)$$

and the difference $|J_{22}^\alpha - J_{33}^\alpha|$ vanishes as shown in Eq. (2.3.10). The nonreciprocity $\Delta\epsilon_\mathbf{k}^\pm$ in the normal state is given by

$$\Delta\epsilon_\mathbf{k}^\pm \equiv \frac{1}{2} (\epsilon_\mathbf{k}^{e\pm} - \epsilon_{-\mathbf{k}}^{e\pm}) = \frac{\mathbf{g}_\mathbf{k} \cdot \mathbf{h}}{\sqrt{g_\mathbf{k}^2 + h^2}} + O((\mathbf{g}_\mathbf{k} \cdot \mathbf{h})^2). \quad (2.3.12)$$

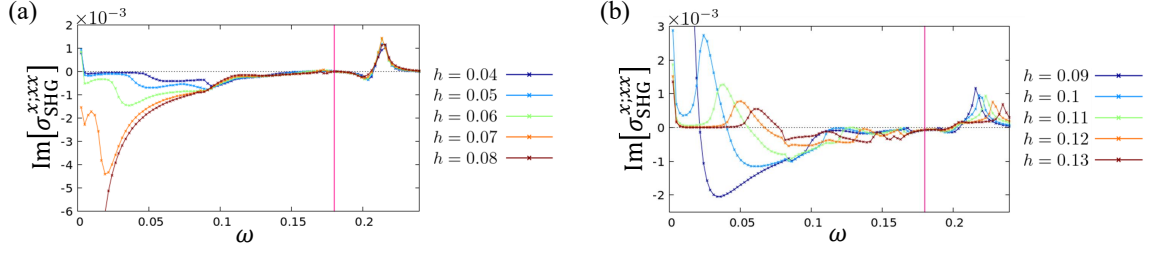


Figure 2.9: The SHG coefficient for various magnitudes of the magnetic field. The unit of the vertical axis is (ÅV^{-2}) . We show $\text{Im}[\sigma_{\text{SHG}}^{x;xx}]$ for (a) $0.04 \leq h \leq 0.08$ and (b) $0.09 \leq h \leq 0.13$. Reproduced from Fig. 15 of Ref. [57]. © 2024 American Physical Society

When we set the magnetic field perpendicular to the plane, the $\mathbf{g}_{\mathbf{k}}$ is normal to the magnetic field in the Rashba system. Consistent with this fact, the $J_{22}^{\alpha} - J_{33}^{\alpha}$ vanishes when we set the angle $\theta = 90^{\circ}$ [Fig. 2.8(c)].

In this section, we have shown the characteristic behavior of the photocurrent conductivity $\sigma_{\text{PC}}^{x;xx}$ around the topological transition. Corresponding to the gap closing, the peak position shifts to a lower frequency with approaching the topological transition. More interestingly, we predicted the sign change of the resonant photocurrent conductivity before and after the topological transition. This sign change is due to the band inversion. Because the occupied band (E_2) and the unoccupied band (E_3) are inverted, the sign of $J_{22}^{\alpha} - J_{33}^{\alpha}$ and that of the magnetic injection current $\sigma_{\text{Minj(B2)}}^{x;xx}$ change. The background of such a close relationship is the fact that the velocity difference is an essential ingredient of the injection current. Therefore, the magnetic injection current, which is dominant in our setup, is sensitive to the band inversion, and in turn, useful for detecting the topological transition. The sign of the bulk photocurrent conductivity has been measured experimentally without electrical contacts [25, 86]. Thus, application to superconductors is highly anticipated.

Finally, we investigate the SHG by focusing on the change at the topological transition. Figure 2.9 shows the imaginary part of the SHG coefficient $\sigma_{\text{SHG}}^{x;xx}$ for magnetic fields (a) $0.04 \leq h \leq 0.08$ (trivial phase) and (b) $0.09 \leq h \leq 0.13$ (topological phase). We see that the sign of the resonant component in the low-frequency region $\omega < 0.1$ is reversed between the trivial and topological superconducting states. Although the mechanism of the sign change has not been elucidated, the SHG can also be an indicator of topological transition. Time-resolved optical measurement can resolve the experimental difficulty of detecting the sign of high harmonic generation. The ultrafast pump-probe spectroscopy accesses the sign of SHG coefficients [28, 29].

2.4 Discussion and summary

We have shown the superconducting nonlinear optical responses in the Rashba s -wave superconductors under the magnetic field. We showed that the magnetic field is essential for the nonlinear responses in broad classes of superconductors. In particular, $\mathcal{T}$ -symmetry breaking nullifies the selection rule forbidding the optical transition between electron and hole bands in $\mathcal{T}$ -symmetric s -wave superconductors. Therefore, the nonlinear responses due to Bogoliubov quasiparticles can occur in conventional superconductors under magnetic fields. In addition, we found a component of superconducting photocurrent generation that emerges under the magnetic field and is closely related to the Dirac point. We ascribe the characteristic dependence on magnetic fields to this component.

The Rashba superconductors have been intensively studied for the realization and detection of topological superconductivity. We showed the sign reversals of photocurrent conductivities and SHG coefficient at the topological transition. Furthermore, we verify that the sign reversal of the photocurrent conductivity occurs associated with the gap closing and band inversion. The longitudinal component of linear conductivity does not show the sign reversal and this characteristic behavior is unique to the nonlinear responses. Therefore, the nonlinear superconducting optics can provide a bulk probe for the detection of topological superconductivity in Rashba systems.

Finally, we comment on the vertex correction for the optical conductivity in the superconductors. In this chapter, we considered the dynamics of the quasiparticle alone and ignored collective-mode excitation. However, incorporating collective-mode excitations is essential to maintain gauge invariance and consistency with the gap equation [49, 81, 87, 88]. In the next chapter, we will present numerical results for photocurrent conductivity that include contributions from the collective mode via the vertex correction. We will show that the sign reversal of the photocurrent conductivity at the topological transition is reproduced even when considering the collective mode excitations.

Chapter 3

Enhancement of nonlinear optical responses in superconductors: collective modes and multiband effects

Collective modes, such as the Higgs mode, are a key ingredient of the dynamics in superconductors. Since the collective modes are closely tied to the Cooper pairs' quantum condensation, they are intriguing phenomena in superconductors. Furthermore, to satisfy conservation laws in superconductors, it is essential to account for collective-mode excitations [89].

In this chapter, we investigate the effect of collective excitation modes on the linear and second-order optical responses based on the self-consistent response approximation, which is formulated using the Kadanoff-Baym method [79]. The formulation accounts for collective modes via the vertex correction. We focus again on the optical responses in *s*-wave superconductors with Rashba spin-orbit coupling and a magnetic field. Our main results reveal that the Higgs mode enhances the optical responses when the Fermi level is close to the Dirac point. The enhancement is due to multiband effects characterized by interband pairing.

We also demonstrate the sign reversal of the photocurrent conductivity around the topological transition with increasing Zeeman field. This finding supports the prediction in the previous chapter without considering collective excitation modes. The sign reversal phenomenon is attributed to the magnetic injection current modified by the Higgs mode, and is proposed as a bulk probe of topological superconductors (TSCs). We also discuss the interplay of quantum geometry and collective modes.

This chapter is organized as follows. In Sec. 3.1, we formulate the self-consistent response approximation for linear and second-order nonlinear responses. In subsequent sections, we study the linear and nonlinear optical conductivities in the Rashba superconductors by using the self-consistent approximation. In Sec. 3.2, we explain the setup for the numerical demonstration and the characteristics of the Rashba superconductors. Then, we present our main results in Secs. 3.3 and 3.4. In Sec. 3.3, we show the linear optical conductivity and discuss the effect of the collective modes and interband pairing. In Sec. 3.4, we investigate the photocurrent conductivity around the topological transition, demonstrating sign reversal and clarifying the role of the collective modes. In Sec. 3.5, we discuss the effect of the long-range Coulomb interaction and material platforms. Finally, we present a summary of this chapter in Sec. 3.6.

3.1 Vertex correction for optical responses in superconductors

In the mean-field formalism, a naive calculation of optical responses may lead to a violation of charge conservation and potentially to unphysical results because of the $U(1)$ -gauge symmetry breaking of the mean field [89]. For the linear response, Nambu resolved this issue by incorporating the Nambu-Coldstone (NG) mode through the vertex correction consistently with the gap equation [49]. After his study, vertex correction has been adopted in the formulation of linear optical responses for a wide range of superconductors [87, 88, 90]. Other various method [91–99] beyond the mean-field formalism have also been proposed to take into account the collective mode. Recent theoretical studies numerically demonstrated non-negligible effects of the vertex correction on the second-order nonlinear responses by the Nambu’s method [81] and the “consistent fluctuations of order parameters” method [82, 83].

Our method for calculating optical responses is based on the self-consistent response approximation. When constructing the self-consistent response approximation, we adopt the Kadanoff-Baym method [100, 101], which maintains the total particle number conservation law. The formula produces physically reasonable results not only for conventional superconductors but also for various unconventional superconductors. The vertex correction incorporates the collective mode excitations into the response functions.

On the other hand, novel collective modes, such as the Leggett mode [50] and Bardasis-Schrieffer mode [52], arise from multi-component Cooper channels. Optical response is a promising probe for detecting these collective modes [102–106]. Thus, we extend the formula for optical responses to account for the multi-component Cooper channels.

In this section, we explain the formulation of the self-consistent response approximation. In Subsect. 3.1.1, we discuss the particle number conservation law in the effective

Hamiltonian with a separable attractive interaction. In Subsect. 3.1.2, the Kadanoff-Baym method is adopted to derive the equation of motion of Green's functions that satisfies the total particle number conservation law. In Subsect. 3.1.3, we formulate the self-consistent response approximation for linear and second-order nonlinear responses.

3.1.1 Particle number conservation law in effective Hamiltonian

The effective Hamiltonian with separable attractive interaction is often used to study unconventional superconductivity without computational difficulties. We can adopt an effective Hamiltonian of general form to study various classes of unconventional superconductivity, such as anisotropic superconductivity, helical superconductivity, and multiband superconductivity. In this subsection, we discuss gauge symmetry and the conservation law of the effective Hamiltonian. The effective Hamiltonian does not always preserve the local gauge symmetry, while the total particle number is conserved.

Effective Hamiltonian

We consider a pairing interaction in momentum space,

$$H_{\text{pair}} = - \sum_{\mathbf{k}, \mathbf{k}', \mathbf{q}} U_{abcd}(\mathbf{k}, \mathbf{k}', \mathbf{q}) C_{\mathbf{k}+\mathbf{q}a}^\dagger C_{-\mathbf{k}+\mathbf{q}b}^\dagger C_{-\mathbf{k}'+\mathbf{q}c} C_{\mathbf{k}'+\mathbf{q}d}, \quad (3.1.1)$$

where $C_{\mathbf{k}a}$ ($C_{\mathbf{k}a}^\dagger$) is the annihilation (creation) operator of electrons. We here reduce the pairing interaction to a simple form which stabilizes superconductivity that we aim to study.

If the total momentum of Cooper pairs is zero, we can omit $\mathbf{q} \neq \mathbf{0}$ terms in Eq.(3.1.1). The pairing interaction for $\mathbf{q} = \mathbf{0}$ can be represented by bases of the irreducible representation Γ with respect to the symmetry of the system [107],

$$U_{abcd}(\mathbf{k}, \mathbf{k}', \mathbf{q}) = \frac{U}{2V} \varphi_{ab}(\mathbf{k}) \varphi_{cd}^\dagger(\mathbf{k}') \delta_{\mathbf{q}, \mathbf{0}}. \quad (3.1.2)$$

When we focus on one of the bases of the pair potential $\varphi_{ab}(\tilde{\Gamma}, \tilde{m}; \mathbf{k}) =: \varphi_{ab}(\mathbf{k})$, we can reduce the pairing interaction,

$$U_{abcd}(\mathbf{k}, \mathbf{k}', \mathbf{q}) = \frac{U}{2V} \varphi_{ab}(\mathbf{k}) \varphi_{cd}^\dagger(\mathbf{k}') \delta_{\mathbf{q}, \mathbf{0}}. \quad (3.1.3)$$

The basis function $\varphi_{ab}(\mathbf{k})$ has the following symmetry: $\varphi_{ab}(\mathbf{k}) = -\varphi_{ba}(-\mathbf{k})$. When we consider Cooper pairs that have a finite total momentum $2\bar{\mathbf{q}}$, we revise the effective pairing interaction,

$$U_{abcd}(\mathbf{k}, \mathbf{k}', \mathbf{q}) = \frac{U}{2V} \varphi_{ab}(\mathbf{k}) \varphi_{cd}^\dagger(\mathbf{k}') \delta_{\mathbf{q}, \bar{\mathbf{q}}}. \quad (3.1.4)$$

By this simplification, the pairing interaction Hamiltonian is written by

$$H_{\text{pair}} = -\frac{U}{2V} \sum_{\mathbf{k}, \mathbf{k}'} \varphi_{ab}(\mathbf{k}) \varphi_{cd}^\dagger(\mathbf{k}') c_{\mathbf{k}a}^\dagger c_{-\mathbf{k}b}^\dagger c_{-\mathbf{k}'c} c_{\mathbf{k}'d}, \quad (3.1.5)$$

where we denote $c_{\mathbf{k}a} \equiv C_{\mathbf{k}+\bar{\mathbf{q}}a}$ for simplicity.

We can deal with the pairing interaction with the mean field approximation. The pair potential corresponding to the pairing symmetry $\varphi_{ab}(\mathbf{k})$ is introduced by

$$\Delta = \frac{U}{V} \sum_{\mathbf{k}} \varphi_{ab}^\dagger(\mathbf{k}) \langle c_{\mathbf{k}b} c_{-\mathbf{k}a} \rangle, \quad (3.1.6)$$

$$\Delta_{ab}(\mathbf{k}) = \varphi_{ab}(\mathbf{k}) \Delta, \quad (3.1.7)$$

approximating the interaction term of Hamiltonian as,

$$\frac{1}{2} \sum_{\mathbf{k}} \left[c_{-\mathbf{k}a} \Delta_{ab}^\dagger(\mathbf{k}) c_{\mathbf{k}b} + c_{\mathbf{k}a}^\dagger \Delta_{ab}(\mathbf{k}) c_{-\mathbf{k}b}^\dagger \right] + \frac{1}{2U} \sum_{\mathbf{k}} |\Delta|^2. \quad (3.1.8)$$

Total particle number conservation law

We here discuss the gauge symmetry and the particle number conservation law in the effective Hamiltonian. We consider the effective Hamiltonian with vector potential

$$H(\tau) = \sum_{\mathbf{k}} \mathcal{H}_{ab}(\mathbf{k}; \mathbf{A}(\tau)) C_{\mathbf{k}a}^\dagger C_{\mathbf{k}b} - \sum_{\mathbf{k}, \mathbf{k}', \mathbf{q}'} U_{abcd}(\mathbf{k}, \mathbf{k}', \mathbf{q}') C_{\mathbf{k}+\mathbf{q}'a}^\dagger C_{-\mathbf{k}+\mathbf{q}'b}^\dagger C_{-\mathbf{k}'+\mathbf{q}'c} C_{\mathbf{k}'+\mathbf{q}'d},$$

where τ is the (imaginary) time and we adopt the velocity gauge $\mathcal{H}_{ab}(\mathbf{k}; \mathbf{A}(\tau)) = \mathcal{H}_{ab}(\mathbf{k} - \mathbf{A}(\tau))$. The effective Hamiltonian has the global gauge symmetry as it is invariant under the gauge transformation $C_{\mathbf{k}a} \rightarrow e^{i\theta} C_{\mathbf{k}a}$. Thus, the total particle number conservation law is derived from the Heisenberg equation

$$\frac{\partial}{\partial \tau} N(\tau) = 0, \quad (3.1.9)$$

where we defined the total particle number N as

$$N = \sum_{\mathbf{k}, i} C_{\mathbf{k}i}^\dagger C_{\mathbf{k}i} = \sum_{\mathbf{k}, i} c_{\mathbf{k}i}^\dagger c_{\mathbf{k}i}. \quad (3.1.10)$$

3.1.2 Equation of motion based on the Kadanoff-Baym method

We consider the imaginary-time evolution of Green's functions using the Kadanoff-Baym method. We verify that the equation of motion derived from the Kadanoff-Baym method satisfies the total particle-number conservation law, whereas the mean-field approximation, in which we consider only the time evolution of Bogoliubov quasiparticles, does not. When

assuming the effective pairing interaction as Eq.(3.1.4), the imaginary time-dependent effective Hamiltonian in Eq.(3.1.9) is approximated as

$$H(\tau) = \sum_{\mathbf{k}} \mathcal{H}_{ab}(\mathbf{k} + \mathbf{q}; \mathbf{A}(\tau)) c_{\mathbf{k}a}^\dagger c_{\mathbf{k}b} + \frac{1}{2} \sum \left[c_{-\mathbf{k}a} \Delta_{ab}^+(\mathbf{k}, \tau; \mathbf{A}) c_{\mathbf{k}b} + c_{\mathbf{k}a}^\dagger \Delta_{ab}(\mathbf{k}, \tau; \mathbf{A}) c_{-\mathbf{k}b}^\dagger \right], \quad (3.1.11)$$

where we ignore the constant term for simplicity and denote the Cooper pairs' total momentum as $2\mathbf{q}$. We introduce the mean field Δ_{ab} as it depends on the imaginary time and the vector potential. The mean field is determined by the Kadanoff-Baym method as we show later.

We introduce the Green's functions ($0 < \tau, \tau' < \beta$) as

$$\mathcal{G}_{ab}(\mathbf{k}, \tau, \tau') = -\langle T_\tau [c_{\mathbf{k}a}(\tau) c_{\mathbf{k}b}^\dagger(\tau')] \rangle_{\text{tot}}, \quad (3.1.12)$$

$$\mathcal{F}_{ab}(\mathbf{k}, \tau, \tau') = -\langle T_\tau [c_{\mathbf{k}a}(\tau) c_{-\mathbf{k}b}(\tau')] \rangle_{\text{tot}}, \quad (3.1.13)$$

$$\mathcal{G}_{ab}^+(\mathbf{k}, \tau, \tau') = -\langle T_\tau [c_{-\mathbf{k}a}^+(\tau) c_{-\mathbf{k}b}(\tau')] \rangle_{\text{tot}}, \quad (3.1.14)$$

$$\mathcal{F}_{ab}^+(\mathbf{k}, \tau, \tau') = -\langle T_\tau [c_{-\mathbf{k}a}^+(\tau) c_{\mathbf{k}b}^\dagger(\tau')] \rangle_{\text{tot}}, \quad (3.1.15)$$

where we adopt the Heisenberg representation

$$c_{\mathbf{k}a}(\tau) = S^+(0, \tau) c_{\mathbf{k}a} S(\tau, 0), \quad (3.1.16)$$

$$c_{\mathbf{k}a}^+(\tau) = S^+(0, \tau) c_{\mathbf{k}a}^\dagger S(\tau, 0), \quad (3.1.17)$$

$$S(\tau, 0) = T_\tau \exp \left[- \int_0^\tau H(\tilde{\tau}) d\tilde{\tau} \right], \quad (3.1.18)$$

$$S^+(0, \tau) = T_\tau^+ \exp \left[\int_0^\tau H(\tilde{\tau}) d\tilde{\tau} \right]. \quad (3.1.19)$$

The notation T_τ (T_τ^+) means the (inverse) imaginary time-ordered product. The expectation value $\langle \cdots \rangle_{\text{tot}}$ is defined as

$$\langle \hat{X} \rangle_{\text{tot}} = \frac{1}{Z[\mathbf{A}]} \text{Tr} \left[\hat{X} S(\beta, 0) \right], \quad Z[\mathbf{A}] = \text{Tr}[S(\beta, 0)]. \quad (3.1.20)$$

The imaginary time evolution of the Green's function is determined by the Heisenberg equation

$$\frac{\partial}{\partial \tau} c_{\mathbf{k}a}(\tau) = [H_H(\tau), c_{\mathbf{k}a}(\tau)], \quad (3.1.21)$$

$$\frac{\partial}{\partial \tau} c_{\mathbf{k}a}^+(\tau) = [H_H(\tau), c_{\mathbf{k}a}^+(\tau)], \quad (3.1.22)$$

$$H_H(\tau) = S^+(0, \tau) H(\tau) S(\tau, 0). \quad (3.1.23)$$

Thus, we derive the equation of motion

$$\begin{aligned} & \left(-\frac{\partial}{\partial \tau} \delta_{aj} - \overleftrightarrow{\mathcal{H}}_{aj}(\mathbf{k}; \mathbf{A}(\tau)) \right) \overleftrightarrow{\mathcal{G}}_{jb}(\mathbf{k}, \tau, \tau'; \mathbf{A}) \\ &= \overleftrightarrow{\mathcal{G}}_{aj}(\mathbf{k}, \tau, \tau'; \mathbf{A}) \left(\frac{\partial}{\partial \tau'} \delta_{aj} - \overleftrightarrow{\mathcal{H}}_{aj}(\mathbf{k}; \mathbf{A}(\tau')) \right) \\ &= \delta(\tau - \tau') \delta_{ab}, \end{aligned} \quad (3.1.24)$$

where we rewrite the Green's function in the Nambu space

$$\overleftrightarrow{\mathcal{G}}_{ab}(\mathbf{k}, \tau, \tau'; \mathbf{A}) = \begin{pmatrix} \mathcal{G}_{ab} & \mathcal{F}_{ab} \\ \mathcal{F}_{ab}^+ & \mathcal{G}_{ab}^+ \end{pmatrix}(\mathbf{k}, \tau, \tau'; \mathbf{A}), \quad (3.1.25)$$

and the Bogoliubov-de Genne (BdG) Hamiltonian

$$\overleftrightarrow{\mathcal{H}}_{ab}(\mathbf{k}, \tau; \mathbf{A}) = \begin{pmatrix} \mathcal{H}_{ab}(\mathbf{k} + \mathbf{q}; \mathbf{A}(\tau)) & \Delta_{ab}(\mathbf{k}, \tau; \mathbf{A}) \\ \Delta_{ab}^+(\mathbf{k}, \tau; \mathbf{A}) & -\mathcal{H}_{ab}^\top(-\mathbf{k} + \mathbf{q}; \mathbf{A}(\tau)) \end{pmatrix}. \quad (3.1.26)$$

We can rewrite Eq. (3.1.24) as

$$\int d\tau_1 \overleftrightarrow{\mathcal{G}}_{aj}^{-1}(\mathbf{k}, \tau, \tau_1; \mathbf{A}) \overleftrightarrow{\mathcal{G}}_{jb}(\mathbf{k}, \tau_1, \tau'; \mathbf{A}) = \delta(\tau - \tau') \delta_{ab}, \quad (3.1.27)$$

$$\int d\tau_1 \overleftrightarrow{\mathcal{G}}_{aj}(\mathbf{k}, \tau, \tau_1; \mathbf{A}) \overleftrightarrow{\mathcal{G}}_{jb}^{-1}(\mathbf{k}, \tau_1, \tau'; \mathbf{A}) = \delta(\tau - \tau') \delta_{ab}, \quad (3.1.28)$$

where we define $\overleftrightarrow{\mathcal{G}}^{-1}$ as

$$\overleftrightarrow{\mathcal{G}}_{ij}^{-1}(\mathbf{k}, \tau, \tau'; \mathbf{A}) = \left\{ \delta_{ij} \frac{\partial}{\partial \tau'} - \overleftrightarrow{\mathcal{H}}_{ij}(\mathbf{k}, \tau'; \mathbf{A}) \right\} \delta(\tau - \tau'). \quad (3.1.29)$$

Next, we consider the time evolution of the total particle number. The expectation value of the total particle number is given by the normal Green's function as

$$\langle N(\tau) \rangle_{\text{tot}} = \sum_{\mathbf{k}, a} \mathcal{G}_{aa}(\mathbf{k}, \tau^-, \tau^+; \mathbf{A}), \quad (3.1.30)$$

where we define $\tau_{\pm} = \tau \pm \delta$ with an infinitesimal positive number $\delta > 0$. Thus, the imaginary time derivative of $\langle N(\tau) \rangle_{\text{tot}}$ is given by

$$\begin{aligned} & \frac{d}{d\tau} \langle N(\tau) \rangle_{\text{tot}} \\ &= \sum_{\mathbf{k}, a} \left[\left(\frac{\partial}{\partial \tau} + \frac{\partial}{\partial \tau'} \right) \mathcal{G}_{aa}(\mathbf{k}, \tau, \tau'; \mathbf{A}) \right]_{\tau=\tau^-, \tau'=\tau^+} \end{aligned} \quad (3.1.31)$$

$$\begin{aligned} &= \sum_{a, j, \mathbf{k}} \mathcal{G}_{aj}(\mathbf{k}, \tau^-, \tau^+; \mathbf{A}) \mathcal{H}_{ja}(\mathbf{k} + \mathbf{q}, \tau; \mathbf{A}) - \sum_{a, j, \mathbf{k}} \mathcal{H}_{aj}(\mathbf{k} + \mathbf{q}, \tau; \mathbf{A}) \mathcal{G}_{ja}(\mathbf{k}, \tau^-, \tau^+; \mathbf{A}) \\ &+ \sum_{a, j, \mathbf{k}} \mathcal{F}_{aj}(\mathbf{k}, \tau^-, \tau^+; \mathbf{A}) \Delta_{ja}^+(\mathbf{k}, \tau; \mathbf{A}) - \sum_{a, j, \mathbf{k}} \Delta_{aj}(\mathbf{k}, \tau; \mathbf{A}) \mathcal{F}_{ja}^+(\mathbf{k}, \tau^-, \tau^+; \mathbf{A}). \end{aligned} \quad (3.1.32)$$

We used Eq. (3.1.24) in the second equation. The first and second terms cancel each other after the summation over the indices a and j , while the third and fourth terms remain. When we only consider the dynamics of Bogoliubov quasiparticles but ignore the imaginary time dependence of the mean field $\Delta_{aj}(\mathbf{k})$, the third and fourth terms can be finite, and the total particle number is not necessarily conserved. Therefore, the imaginary time dependence of the mean field is essential for the total particle number conservation in the superconducting state.

We then assume that the imaginary time dependence of the mean field is determined by the Kadanoff-Baym method,

$$\Delta_{ab}^+(\mathbf{k}, \tau; \mathbf{A}) = -\frac{U}{V} \varphi_{ab}^\dagger(\mathbf{k}) \sum_{\mathbf{k}', c, d} \varphi_{dc}(\mathbf{k}') \mathcal{F}_{cd}^+(\mathbf{k}', \tau^+, \tau^-; \mathbf{A}), \quad (3.1.33)$$

$$\Delta_{ab}(\mathbf{k}, \tau; \mathbf{A}) = -\frac{U}{V} \varphi_{ab}(\mathbf{k}) \sum_{\mathbf{k}', c, d} \varphi_{dc}^\dagger(\mathbf{k}') \mathcal{F}_{cd}(\mathbf{k}', \tau^+, \tau^-; \mathbf{A}). \quad (3.1.34)$$

The third and fourth terms in Eq. (3.1.32) are rewritten as

$$\begin{aligned} & \sum \mathcal{F}_{aj}(\mathbf{k}, \tau^-, \tau^+; \mathbf{A}) \Delta_{ja}^+(\mathbf{k}, \tau; \mathbf{A}) \\ &= -\frac{U}{V} \left[\sum \mathcal{F}_{aj}(\mathbf{k}, \tau^-, \tau^+; \mathbf{A}) \varphi_{ja}^\dagger(\mathbf{k}) \right] \left[\sum \varphi_{dc}(\mathbf{k}') \mathcal{F}_{cd}^+(\mathbf{k}', \tau^+, \tau^-; \mathbf{A}) \right], \end{aligned} \quad (3.1.35)$$

$$\begin{aligned} & \sum \Delta_{aj}(\mathbf{k}, \tau; \mathbf{A}) \mathcal{F}_{ja}^+(\mathbf{k}, \tau^-, \tau^+; \mathbf{A}) \\ &= -\frac{U}{V} \left[\sum \varphi_{dc}^\dagger(\mathbf{k}') \mathcal{F}_{cd}(\mathbf{k}', \tau^+, \tau^-; \mathbf{A}) \right] \left[\sum \varphi_{aj}(\mathbf{k}) \mathcal{F}_{ja}^+(\mathbf{k}, \tau^-, \tau^+; \mathbf{A}) \right]. \end{aligned} \quad (3.1.36)$$

Using the relation $\mathcal{F}_{ab}(\mathbf{k}, \tau^-, \tau^+; \mathbf{A}) = \mathcal{F}_{ab}(\mathbf{k}, \tau^+, \tau^-; \mathbf{A})$, we verify that the third and fourth terms in Eq. (3.1.32) cancel each other and the total particle number conservation law $\frac{d}{d\tau} \langle N(\tau) \rangle_{\text{tot}} = 0$ is satisfied. We can also obtain the standard gap equation for superconductivity from the assumptions (3.1.35) and (3.1.36) when the external electric field is absent ($\mathbf{A} = \mathbf{0}$). Thus, Eqs. (3.1.35) and (3.1.36) are expected to be appropriate as the mean-field pair potential under the external electric field. In the following subsection, we derive the formula of the self-consistent response approximation from Eqs. (3.1.27) and (3.1.28).

3.1.3 Self-consistent response approximation

We demonstrate that the total particle number conservation law is satisfied when we apply the Kadanoff-Baym method and consider the imaginary time dependence of the mean-field pair potential. Therefore, the self-consistent response approximation formulated below, which is derived from the equation of motion based on the Kadanoff-Baym method, is physically reasonable. Because the Green's function contains the effect of electric fields

nonperturbatively, we can derive the formulas of linear and nonlinear responses that satisfy the total number conservation in any order of the electric field expansion. This subsection shows the derivation of the linear and second-order nonlinear response formulas based on the self-consistent response approximation. The vertex correction is taken into account through the electric field dependence of the mean-field pair potential.

The time evolution of the electric current is formulated by the equation of motion based on the Kadanoff-Baym method. Following statistical mechanical relations, we derive the electric current from varying a parameter in the grand partition function. In the Luttinger-Ward approach [108, 109], $\ln Z$ is evaluated as an integral of the potential energy with respect to the coupling constant parameter λ ,

$$\ln Z = \text{tr} \ln[-\overleftrightarrow{\mathcal{G}}_0] + \int_0^1 \frac{d\lambda}{2\lambda} \text{tr} \overleftrightarrow{\Sigma}_\lambda \overleftrightarrow{\mathcal{G}}_\lambda, \quad (3.1.37)$$

where $\overleftrightarrow{\Sigma}_\lambda$ and $\overleftrightarrow{\mathcal{G}}_\lambda$ are the self-energy and the Green's function obtained with replacing the coupling constant $U \rightarrow \lambda U$.

Next, we derive the electric current by considering the change of $\ln Z$ with varying the vector potential $\mathbf{A}(\tau)$ in $\mathcal{H}_{ab}(\mathbf{k} + \mathbf{q}; \mathbf{A}(\tau))$,

$$\langle J^\alpha(\tau; \mathbf{A}) \rangle = \frac{\partial \ln Z[\mathbf{A}]}{\partial A^\alpha(\tau)} = \frac{1}{2} \sum \text{Tr} \left[\overleftrightarrow{J}_{ab}^\alpha(\mathbf{k}; \mathbf{A}(\tau)) \overleftrightarrow{\mathcal{G}}_{ba}(\mathbf{k}, \tau^-, \tau^+; \mathbf{A}) \right]. \quad (3.1.38)$$

We introduced the current operator,

$$\overleftrightarrow{J}_{ab}^{\alpha_1 \dots \alpha_n}(\mathbf{k}; \mathbf{A}(\tau)) = (-1)^n \left. \frac{\partial^n \overleftrightarrow{\mathcal{H}}_{ab}^{(0)}(\mathbf{k}; \mathbf{A})}{\partial A^{\alpha_1} \dots \partial A^{\alpha_n}} \right|_{\mathbf{A}=\mathbf{A}(\tau)}, \quad (3.1.39)$$

with use of the following notation,

$$\overleftrightarrow{\mathcal{H}}_{ab}^{(0)}(\mathbf{k}; \mathbf{A}) = \begin{pmatrix} \mathcal{H}_{ab}(\mathbf{k} + \mathbf{q}; \mathbf{A}) & 0 \\ 0 & -\mathcal{H}_{ab}^\top(-\mathbf{k} + \mathbf{q}; \mathbf{A}) \end{pmatrix}. \quad (3.1.40)$$

It should be notice that the electric current $\langle J^\alpha(\tau; \mathbf{A}) \rangle$ depends on the vector potential through the explicit current operator's dependence on the instant vector potential $\mathbf{A}(\tau)$ and through the implicit dependence of the Green's function on the imaginary-time history of $\mathbf{A}(\tau)$.

Linear response

First, we formulate the self-consistent response approximation for the linear response functions. The vertex correction corresponding to the ladder approximation appears because of the dependence of the mean-field pair potential on the vector potential.

From the vector potential dependence of the electric current $\langle J^\alpha(\tau; \mathbf{A}) \rangle$, the imaginary-time linear response function $\tilde{\chi}^{\alpha\beta}$ is defined as

$$\tilde{\chi}^{\alpha\beta}(\tau, \tau') = \frac{\delta \langle J^\alpha(\tau; \mathbf{A}) \rangle}{\delta A^\beta(\tau')}. \quad (3.1.41)$$

Here, we omit the index a, b and rewrite the electric current for simplicity,

$$\langle J^\alpha(\mathbf{A}(\tau)) \rangle = \frac{1}{2} \text{Tr}[J^\alpha(\tau) \mathcal{G}(\tau^-, \tau^+)]. \quad (3.1.42)$$

From Eqs. (3.1.27) and (3.1.28), we obtain the relation

$$\begin{aligned} \frac{\delta \mathcal{G}(\tau, \tau')}{\delta A^\alpha(\bar{\tau}_1)} &= - \int d\tau_1 d\tau_2 \mathcal{G}(\tau, \tau_1) \frac{\delta \mathcal{G}^{-1}(\tau_1, \tau_2)}{\delta A^\alpha(\bar{\tau}_1)} \mathcal{G}(\tau_2, \tau') \\ &= - \int d\tau_1 d\tau_2 \mathcal{G}(\tau, \tau_1) \Gamma^\alpha(\tau_1, \tau_2; \bar{\tau}_1) \mathcal{G}(\tau_2, \tau'), \end{aligned} \quad (3.1.43)$$

where the vertex function Γ^α is introduced as

$$\Gamma^\alpha(\tau_1, \tau_2; \bar{\tau}_1) \equiv \frac{\delta \mathcal{G}^{-1}(\tau_1, \tau_2)}{\delta A^\alpha(\bar{\tau}_1)}. \quad (3.1.44)$$

Thus, the linear response function $\tilde{\chi}^{\alpha\beta}$ is represented with the Green's function and the vertex function as

$$\begin{aligned} \tilde{\chi}^{\alpha\beta}(\tau, \tau') &= - \frac{1}{2} \sum \text{Tr} \left[\overleftrightarrow{J}_{ab}^{\alpha\beta}(\mathbf{k}) \overleftrightarrow{\mathcal{G}}_{ba}(\mathbf{k}', \tau^-, \tau^+) \delta(\tau - \tau') \right] \\ &\quad - \frac{1}{2} \sum \int d\tau_1 \text{Tr} \left[\overleftrightarrow{J}_{ab}^\alpha(\mathbf{k}) \overleftrightarrow{\mathcal{G}}_{bi}(\mathbf{k}, \tau, \tau_1) \overleftrightarrow{\Gamma}_{ij}^\beta(\mathbf{k}, \tau_1; \tau') \overleftrightarrow{\mathcal{G}}_{ja}(\mathbf{k}, \tau_1, \tau) \right]. \end{aligned} \quad (3.1.45)$$

We then show a self-consistent equation that determines the vertex function. By the definition of the vertex function Eq. (3.1.44), we can simplify the imaginary-time dependence of the vertex function as

$$\overleftrightarrow{\Gamma}_{ab}^\alpha(\mathbf{k}, \tau_1, \tau_2; \tau') = \left[\overleftrightarrow{J}_{ab}^\alpha(\mathbf{k}) \delta(\tau_2 - \tau') - \frac{\delta \overleftrightarrow{\Delta}_{ab}(\mathbf{k}, \tau_2; \mathbf{A})}{\delta A^\alpha(\tau')} \right] \delta(\tau_1 - \tau_2) \quad (3.1.46)$$

$$= \overleftrightarrow{\Gamma}_{ab}^\alpha(\mathbf{k}, \tau_2; \tau') \delta(\tau_1 - \tau_2). \quad (3.1.47)$$

In the last line, a simplified notation of the vertex function is introduced and the τ_1 -dependence is omitted. We define $\overleftrightarrow{\Delta}$ as

$$\overleftrightarrow{\Delta}_{ab}(\mathbf{k}, \tau; \mathbf{A}) = \begin{pmatrix} 0 & \Delta_{ab} \\ \Delta_{ab}^+ & 0 \end{pmatrix} (\mathbf{k}, \tau; \mathbf{A}). \quad (3.1.48)$$

It can be rewritten as

$$\begin{aligned} \overleftrightarrow{\Delta}_{ab}(\mathbf{k}, \tau; \mathbf{A}) &= - \frac{U}{V} \varphi_{ab}(\mathbf{k}) \sum \varphi_{dc}^\dagger(\mathbf{k}') \overleftrightarrow{\mathcal{P}}_+^\dagger \overleftrightarrow{\mathcal{G}}_{cd}(\mathbf{k}', \tau^+, \tau^-; \mathbf{A}) \overleftrightarrow{\mathcal{P}}_-^\dagger \\ &\quad - \frac{U}{V} \varphi_{ab}^\dagger(\mathbf{k}) \sum \varphi_{dc}(\mathbf{k}') \overleftrightarrow{\mathcal{P}}_-^\dagger \overleftrightarrow{\mathcal{G}}_{cd}(\mathbf{k}', \tau^+, \tau^-; \mathbf{A}) \overleftrightarrow{\mathcal{P}}_+^\dagger, \end{aligned} \quad (3.1.49)$$

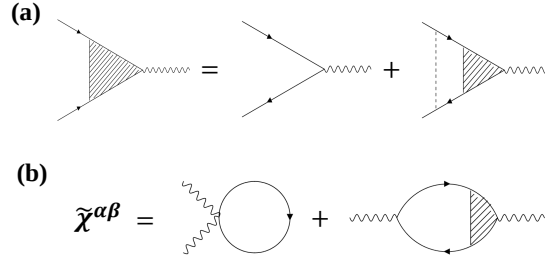


Figure 3.1: Feynman diagram of (a) the self-consistent equation for the vertex function and (b) the linear response function $\tilde{\chi}^{\alpha\beta}$.

where we use the matrices in the Nambu space such that

$$\overleftrightarrow{\mathbf{P}}_+ = \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix}, \quad \overleftrightarrow{\mathbf{P}}_- = \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix}. \quad (3.1.50)$$

The mean-field pair potential $\overleftrightarrow{\Delta}$ depends on the vector potential only through the Green's function. We see that the vector potential derivative of the mean-field pair potential contributes to the vertex function in Eq. (3.1.46). The self-consistent equation of the vertex function is obtained from Eqs. (3.1.43), (3.1.47), and (3.1.49).

We can simply describe the self-consistent equation by introducing the superoperator $\overleftrightarrow{\mathcal{U}}_{ab}(\mathbf{k}, \tau_1)[X]$ acting on $X \equiv \overleftrightarrow{X}_{ab}(\mathbf{k}, \tau)$ as,

$$\overleftrightarrow{\Gamma}_{ab}^\alpha(\mathbf{k}, \tau_1; \tau') = \overleftrightarrow{\mathcal{J}}_{ab}^\alpha(\mathbf{k})\delta(\tau_1 - \tau') + \overleftrightarrow{\mathcal{U}}_{ab}(\mathbf{k}, \tau_1)[\Gamma^\alpha(\tau')], \quad (3.1.51)$$

where the superoperator $\overleftrightarrow{\mathcal{U}}_{ab}(\mathbf{k}, \tau_1)[X]$ is defined as

$$\begin{aligned} & \overleftrightarrow{\mathcal{U}}_{ab}(\mathbf{k}, \tau_1)[X] \\ & \equiv -\frac{U}{V}\varphi_{ab}(\mathbf{k}) \sum \varphi_{dc}^\dagger(\mathbf{k}') \overleftrightarrow{\mathbf{P}}_+ \int d\bar{\tau}_1 \overleftrightarrow{\mathcal{G}}_{ci}(\mathbf{k}', \tau_1, \bar{\tau}_1) \overleftrightarrow{X}_{ij}(\mathbf{k}', \bar{\tau}_1) \overleftrightarrow{\mathcal{G}}_{jd}(\mathbf{k}', \bar{\tau}_1, \tau_1) \overleftrightarrow{\mathbf{P}}_- \\ & \quad - \frac{U}{V}\varphi_{ab}^\dagger(\mathbf{k}) \sum \varphi_{dc}(\mathbf{k}') \overleftrightarrow{\mathbf{P}}_- \int d\bar{\tau}_1 \overleftrightarrow{\mathcal{G}}_{ci}(\mathbf{k}', \tau_1, \bar{\tau}_1) \overleftrightarrow{X}_{ij}(\mathbf{k}', \bar{\tau}_1) \overleftrightarrow{\mathcal{G}}_{jd}(\mathbf{k}', \bar{\tau}_1, \tau_1) \overleftrightarrow{\mathbf{P}}_+. \end{aligned} \quad (3.1.52)$$

The Feynman diagram representing the self-consistent equation is depicted in Fig. 3.1(a).

We then introduce superoperator that transforms the bare electric current operator to the vertex function. We consider a self-consistent equation of an operator Z ,

$$\overleftrightarrow{Z}_{ab}(\mathbf{k}, \tau) = \overleftrightarrow{X}_{ab}(\mathbf{k}, \tau) + \overleftrightarrow{\mathcal{U}}_{ab}(\mathbf{k}, \tau)[Z]. \quad (3.1.53)$$

When the solution Z of the equation is uniquely determined, the Z can be regarded as a function of X such that

$$\overleftrightarrow{Z}_{ab}(\mathbf{k}, \tau) = \overleftrightarrow{Z}_{ab}(\mathbf{k}, \tau)[X]. \quad (3.1.54)$$

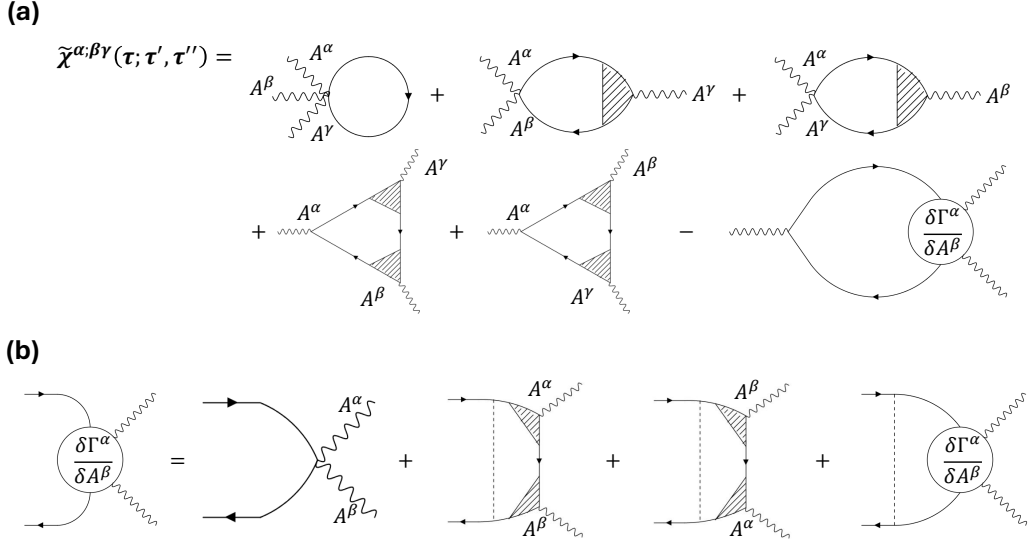


Figure 3.2: Feynman diagram of (a) the second-order nonlinear response function $\tilde{\chi}^{\alpha;\beta\gamma}$ and (b) the self-consistent equation for the two-photon vertex.

Thus, the function $Z[X]$ is a superoperator, and the self-consistent equation of the vertex function is rewritten as

$$\overleftrightarrow{\Gamma}_{ab}^{\alpha}(\mathbf{k}, \tau_1; \tau') = \overleftrightarrow{Z}_{ab}(\mathbf{k}, \tau_1)[J^{\alpha}\delta(\tau_1 - \tau')]. \quad (3.1.55)$$

Finally, we summarize the self-consistent response approximation for the linear response function. The imaginary-time linear response function $\tilde{\chi}^{\alpha\beta}$ is given by Eq. (3.1.45). The diagrammatic illustration of this formula is shown in Fig. 3.1(b). The vertex function Γ is determined by the self-consistent equation [Fig. 3.1(a)], which corresponds to the ladder approximation. We can get the linear response function in the Matsubara frequency presentation $\tilde{\chi}^{\alpha\beta}(i\omega, i\omega')$ after the Fourier transformation with respect to the imaginary time. We introduce a simplified notation of the linear response function,

$$\tilde{\chi}^{\alpha\beta}(i\omega, i\omega') = \tilde{\chi}^{\alpha\beta}(i\omega)\beta\delta(\omega - \omega'), \quad (3.1.56)$$

where we use the imaginary time translation symmetry to omit the ω' -dependence.

Nonlinear response

Next, we derive the self-consistent response approximation for the second-order nonlinear responses. The second-order nonlinear response function $\tilde{\chi}^{\alpha;\beta\gamma}$ is defined as

$$\tilde{\chi}^{\alpha;\beta\gamma}(\tau; \tau', \tau'') \equiv \frac{1}{2!} \frac{\delta^2 \langle J^{\alpha}(\tau) \rangle}{\delta A^{\beta}(\tau') \delta A^{\gamma}(\tau'')}. \quad (3.1.57)$$

The nonlinear response function is related to the nonlinear conductivity $\sigma^{\alpha;\beta\gamma}(\omega; \omega', \omega'')$ as we discuss later. We differentiate Eq. (3.1.43) with the vector potential and obtain the following relation:

$$\begin{aligned} \frac{\delta^2 \mathcal{G}(\tau_1, \tau_2)}{\delta A^\alpha(\tau) \delta A^\beta(\tau')} &= \int d\bar{\tau}_1 d\bar{\tau}_2 \mathcal{G}(\tau_1, \bar{\tau}_1) \Gamma^\alpha(\bar{\tau}_1; \tau) \mathcal{G}(\bar{\tau}_1, \bar{\tau}_2) \Gamma^\beta(\bar{\tau}_2; \tau') \mathcal{G}(\bar{\tau}_2, \tau_2) \\ &\quad + \int d\bar{\tau}_1 d\bar{\tau}_2 \mathcal{G}(\tau_1, \bar{\tau}_1) \Gamma^\beta(\bar{\tau}_1; \tau') \mathcal{G}(\bar{\tau}_1, \bar{\tau}_2) \Gamma^\alpha(\bar{\tau}_2; \tau) \mathcal{G}(\bar{\tau}_2, \tau_2) \\ &\quad - \int d\bar{\tau}_1 \mathcal{G}(\tau_1, \bar{\tau}_1) \frac{\delta \Gamma^\alpha(\bar{\tau}_1; \tau)}{\delta A^\beta(\tau')} \mathcal{G}(\bar{\tau}_1, \tau_2). \end{aligned} \quad (3.1.58)$$

Therefore, the nonlinear response function is derived as

$$\begin{aligned} \tilde{\chi}^{\alpha;\beta\gamma}(\tau; \tau', \tau'') &= \frac{1}{4} \text{Tr} \left[J^{\alpha\beta\gamma} \mathcal{G}(\tau^-, \tau^+) \right] \delta(\tau - \tau') \delta(\tau - \tau'') \\ &\quad + \frac{1}{4} \int d\tau_1 \text{Tr} \left[J^{\alpha\beta} \mathcal{G}(\tau, \tau_1) \Gamma^\gamma(\tau_1; \tau'') \mathcal{G}(\tau_1, \tau) \right] \delta(\tau - \tau') \\ &\quad + \frac{1}{4} \int d\tau_1 \text{Tr} \left[J^{\alpha\gamma} \mathcal{G}(\tau, \tau_1) \Gamma^\beta(\tau_1; \tau') \mathcal{G}(\tau_1, \tau) \right] \delta(\tau - \tau'') \\ &\quad + \frac{1}{4} \int d\tau_1 d\tau_2 \text{Tr} \left[J^\alpha \mathcal{G}(\tau, \tau_1) \Gamma^\beta(\tau_1; \tau') \mathcal{G}(\tau_1, \tau_2) \Gamma^\gamma(\tau_2; \tau'') \mathcal{G}(\tau_2, \tau) \right] \\ &\quad + \frac{1}{4} \int d\tau_1 d\tau_2 \text{Tr} \left[J^\alpha \mathcal{G}(\tau, \tau_1) \Gamma^\gamma(\tau_1; \tau'') \mathcal{G}(\tau_1, \tau_2) \Gamma^\beta(\tau_2; \tau') \mathcal{G}(\tau_2, \tau) \right] \\ &\quad - \frac{1}{4} \int d\tau_1 \text{Tr} \left[J^\alpha \mathcal{G}(\tau, \tau_1) \frac{\delta \Gamma^\beta(\tau_1; \tau')}{\delta A^\gamma(\tau'')} \mathcal{G}(\tau_1, \tau) \right]. \end{aligned} \quad (3.1.59)$$

We diagrammatically show the nonlinear response function in Fig. 3.2(a). The last term contains $\delta \Gamma^\beta(\tau_1; \tau') / \delta A^\gamma(\tau'')$ that is known as the two-photon vertex. The self-consistent equation for the two-photon vertex is depicted in Fig. 3.2(b), which is derived by differentiating the self-consistent equation for the one-photon vertex [Eq. (3.1.51)]. We can write the two-photon vertex by utilizing the superoperator Z ,

$$\begin{aligned} \frac{\delta \overleftrightarrow{\Gamma}_{ab}^\alpha(\mathbf{k}, \tau_1; \tau)}{\delta A^\beta(\tau')} &= \overleftrightarrow{Z}_{ab}(\mathbf{k}, \tau_1) \left[\int d\bar{\tau} \left\{ -U(\tau_1, \bar{\tau}) [\Gamma^\beta(\tau_1; \tau') \mathcal{G}(\tau_1, \bar{\tau}) \Gamma^\alpha(\bar{\tau}; \tau)] \right\} \delta(\tau_1 - \bar{\tau}) \right] \\ &\quad + \overleftrightarrow{Z}_{ab}(\mathbf{k}, \tau_1) \left[\int d\bar{\tau} \left\{ -U(\tau_1, \bar{\tau}) [\Gamma^\alpha(\tau_1; \tau) \mathcal{G}(\tau_1, \bar{\tau}) \Gamma^\beta(\bar{\tau}; \tau')] \right\} \delta(\tau_1 - \bar{\tau}) \right] \\ &\quad + \overleftrightarrow{Z}_{ab}(\mathbf{k}, \tau_1) \left[-J^{\alpha\beta} \delta(\tau_1 - \tau) \delta(\tau_1 - \tau') \right]. \end{aligned} \quad (3.1.60)$$

Thus, we obtain the self-consistent equations for the one-photon and two-photon vertices. By solving the self-consistent equations, we can evaluate the vertices and calculate the nonlinear response function $\tilde{\chi}^{\alpha;\beta\gamma}$ [Eq. (3.1.59)].

We can rewrite the formula of the second-order nonlinear response function in a symmetric form,

$$\begin{aligned} \tilde{\chi}^{\alpha;\beta\gamma}(\tau; \tau', \tau'') &= \frac{1}{2} \tilde{\chi}^{[\alpha\beta\gamma]}(\tau; \tau', \tau'') + \frac{1}{2} \tilde{\chi}^{[\alpha][\beta\gamma]}(\tau; \tau', \tau'') \\ &\quad + \tilde{\chi}^{[\alpha][\beta][\gamma]}(\tau; \tau', \tau'') + \tilde{\chi}^{[\alpha\beta][\gamma]}(\tau; \tau', \tau'') + (\beta \leftrightarrow \gamma, \quad \tau' \leftrightarrow \tau''), \end{aligned} \quad (3.1.61)$$

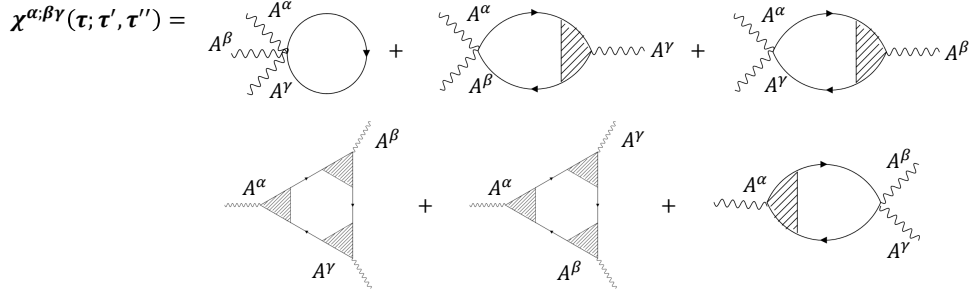


Figure 3.3: Feynman diagrams that contribute to the second-order nonlinear response function $\tilde{\chi}^{\alpha;\beta\gamma}$.

where we define the terms $\tilde{\chi}^{\alpha;\beta\gamma}(\dots)$ as

$$\tilde{\chi}^{\alpha;\beta\gamma}(\tau; \tau', \tau'') = \frac{1}{4} \sum \text{Tr} \left[\overleftrightarrow{J}_{ab}^{\alpha\beta\gamma}(\mathbf{k}) \overleftrightarrow{\mathcal{G}}_{ba}(\mathbf{k}, \tau^-, \tau^+) \right] \delta(\tau' - \tau') \delta(\tau' - \tau''), \quad (3.1.62)$$

$$\begin{aligned} & \tilde{\chi}^{[\alpha][\beta\gamma]}(\tau; \tau', \tau'') \\ &= \frac{1}{4} \int d\tau_1 \sum \text{Tr} \left[\overleftrightarrow{\Gamma}_{ab}^{\alpha}(\mathbf{k}, \tau_1; \tau) \overleftrightarrow{\mathcal{G}}_{bi}(\mathbf{k}, \tau_1, \tau') \overleftrightarrow{J}_{ij}^{\beta\gamma}(\mathbf{k}) \overleftrightarrow{\mathcal{G}}_{ja}(\mathbf{k}, \tau', \tau_1) \right] \delta(\tau' - \tau''), \end{aligned} \quad (3.1.63)$$

$$\begin{aligned} \tilde{\chi}^{[\alpha][\beta][\gamma]}(\tau; \tau', \tau'') &= \frac{1}{4} \int d\tau_1 d\tau_2 d\tau_3 \sum \text{Tr} \left[\overleftrightarrow{\Gamma}_{ab}^{\alpha}(\mathbf{k}, \tau_1; \tau) \overleftrightarrow{\mathcal{G}}_{bi}(\mathbf{k}, \tau_1, \tau_2) \overleftrightarrow{\Gamma}_{ij}^{\gamma}(\mathbf{k}, \tau_2; \tau'') \right. \\ & \quad \left. \times \mathcal{G}_{jk}(\mathbf{k}, \tau_2, \tau_3) \overleftrightarrow{\Gamma}_{kl}^{\beta}(\mathbf{k}, \tau_3; \tau') \overleftrightarrow{\mathcal{G}}_{la}(\mathbf{k}, \tau_3, \tau_1) \right], \end{aligned} \quad (3.1.64)$$

$$\begin{aligned} & \tilde{\chi}^{[\alpha\beta][\gamma]}(\tau; \tau', \tau'') \\ &= \frac{1}{4} \int d\tau_1 \sum \text{Tr} \left[\overleftrightarrow{J}_{ab}^{\alpha\beta}(\mathbf{k}) \overleftrightarrow{\mathcal{G}}_{bi}(\mathbf{k}, \tau, \tau_1) \overleftrightarrow{\Gamma}_{ij}^{\gamma}(\mathbf{k}, \tau_1; \tau'') \overleftrightarrow{\mathcal{G}}_{ja}(\mathbf{k}, \tau_1, \tau) \right] \delta(\tau - \tau'). \end{aligned} \quad (3.1.65)$$

Note that the one-photon vertex function is obtained by solving the self-consistent equation, Eq. (3.1.55). The detailed derivation of the symmetric form is presented in Appendix A. It is beneficial to rewrite the formula for the nonlinear current response function in a symmetric form, as illustrated in Fig. 3.3. Because the symmetric form does not contain the two-photon vertex explicitly, we do not have to solve the self-consistent equation for the two-photon vertex, and the computational task is significantly reduced. We get the nonlinear response function in the Matsubara frequency presentation $\tilde{\chi}^{\alpha;\beta\gamma}(i\omega; i\omega', i\omega'')$ after the Fourier transformation with respect to the imaginary time. Utilizing the imaginary time translation symmetry, we can simplify the nonlinear response function as

$$\tilde{\chi}^{\alpha;\beta\gamma}(i\omega; i\omega', i\omega'') = \tilde{\chi}^{\alpha;\beta\gamma}(i\omega', i\omega'') \beta \delta(\omega - \omega' - \omega''). \quad (3.1.66)$$

Analytic continuation of nonlinear response function

To discuss the nonlinear optical responses in real time, we need the retarded nonlinear response functions. It is known that the real frequency representation of the retarded nonlinear response functions can be obtained by the analytic continuation of the response functions in the Matsubara frequency representation [1, 110]. We review the analytic continuation of the response functions.

We consider the time evolution of the physical quantity $\hat{A}(t)$ under the external field $F(t)$ and assume a perturbative Hamiltonian

$$\hat{V}(t) = \sum \hat{B}_i(t)F_i(t) + \sum \hat{C}_{ij}(t)F_i(t)F_j(t) + \cdots, \quad (3.1.67)$$

where the operators are given in the interaction representation. The expectation value of $\hat{A}(t)$ is calculated by the density matrix,

$$A(t) = \text{Tr} [\hat{\rho}(t)\hat{A}(t)], \quad (3.1.68)$$

and it is expanded with respect to the external fields,

$$\begin{aligned} A(t) = & \langle \hat{A}(t) \rangle + \sum_i \int_{-\infty}^{\infty} dt_1 \chi_{AB_i}(t_1) F_i(t-t_1) \\ & + \sum_{i,j} \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2 \chi_{AB_i B_j}(t_1, t_2) F_i(t-t_1) F_j(t-t_2) \\ & + \sum_{i,j} \int_{-\infty}^{\infty} dt_1 \chi_{AC_{ij}}(t_1) F_i(t-t_1) F_j(t-t_1) + \cdots. \end{aligned} \quad (3.1.69)$$

The response functions are defined as

$$\chi_{AB_i}(t_1) = i\Theta(t_1) \left\langle \left[\Delta \hat{B}_i(-t_1), \Delta \hat{A}(0) \right] \right\rangle, \quad (3.1.70)$$

$$\chi_{AB_i B_j}(t_1, t_2) = \frac{i^2}{2!} \sum_{\mathcal{P}} \Theta(t_2 - t_1) \Theta(t_1) \left\langle \left[\Delta \hat{B}_j(-t_2), \left[\Delta \hat{B}_i(-t_1), \Delta \hat{A}(0) \right] \right] \right\rangle, \quad (3.1.71)$$

$$\chi_{AC_{ij}}(t_1) = \frac{i}{2!} \sum_{\mathcal{P}} \Theta(t_1) \left\langle \left[\Delta \hat{C}_{ij}(-t_1), \Delta \hat{A}(0) \right] \right\rangle. \quad (3.1.72)$$

The notation $\langle \cdots \rangle$ describes the expectation value under the canonical distribution of an unperturbed Hamiltonian, and $\Delta \hat{X}(t) \equiv \hat{X}(t) - \langle \hat{X}(t) \rangle$. The symbol $\sum_{\mathcal{P}}$ stands for summation over all permutations among the dummy variables such as (B_i, t_1) and (B_j, t_2) .

The linear and nonlinear response functions of the electric current are defined by

$$\begin{aligned} J^\alpha(t) = & \langle \hat{J}^\alpha(t) \rangle + \sum_{\beta} \int_{-\infty}^{\infty} dt_1 \chi^{\alpha\beta}(t_1) A^\beta(t-t_1) \\ & + \sum_{\beta, \gamma} \int_{-\infty}^{\infty} dt_1 dt_2 \chi^{\alpha; \beta\gamma}(t_1, t_2) A^\beta(t-t_1) A^\gamma(t-t_2) + \cdots. \end{aligned} \quad (3.1.73)$$

We can calculate the current response functions $\chi^{\alpha\beta}(t_1)$ and $\chi^{\alpha;\beta\gamma}(t_1, t_2)$ by replacing the operators $\hat{A}$, $\hat{B}$, and $\hat{C}$ in the previous paragraph with $\hat{J}^\alpha$, $\hat{J}^{\alpha\beta}$, and $\hat{J}^{\alpha\beta\gamma}$. We proceed to review the general formulation and later discuss the current responses.

Here, we consider the three-body response function $\chi_{AB_iB_j}(t_1, t_2)$ and discuss the relationship with the counterpart in the imaginary time representation. First, we perform the Fourier transformation with respect to the real time t_i ,

$$\chi_{AB_iB_j}(\omega_1, \omega_2) = \frac{1}{2!} \sum_{\mathcal{P}} \sum_{\lambda_1 \lambda_2 \lambda_3} \frac{\Delta A_{\lambda_1 \lambda_3} \Delta B_{j, \lambda_3 \lambda_2} \Delta B_{i, \lambda_2 \lambda_1}}{\omega_1 + \omega_2 + E_{\lambda_1 \lambda_3} + i(\eta_1 + \eta_2)} \times \left[\frac{P_{\lambda_1 \lambda_2}}{\omega_1 + E_{\lambda_1 \lambda_2} + i\eta_1} - \frac{P_{\lambda_2 \lambda_3}}{\omega_2 + E_{\lambda_2 \lambda_3} + i\eta_2} \right], \quad (3.1.74)$$

where $E_{\lambda_1 \lambda_2} = E_{\lambda_1} - E_{\lambda_2}$, $P_{\lambda_1 \lambda_2} = P_{\lambda_1} - P_{\lambda_2}$, $\Delta A_{\lambda_1 \lambda_3} = \langle \Lambda_1 | \Delta \hat{A}(0) | \Lambda_3 \rangle$, and $P_{\lambda_1} = e^{-\beta E_{\lambda_1}} / \sum_{\lambda} e^{-\beta E_{\lambda}}$. The infinitesimal positive parameters η_1 and η_2 appear owing to the causality of the response functions. Next, we derive the spectral representation of the response functions in the Matsubara frequency domain. The imaginary time evolution of the expectation value $\langle A(\tau) \rangle_{\text{tot}}$ is defined as Eqs. (3.1.16-3.1.20), and we have

$$\begin{aligned} \langle A(\tau) \rangle_{\text{tot}} &= \langle \hat{A}(\tau) \rangle + \sum_i \int_0^\beta d\tau_1 \tilde{\chi}_{AB_i}(\tau_1) F_i(\tau - \tau_1) \\ &\quad + \sum_{i,j} \int_0^\beta d\tau_1 \int_0^\beta d\tau_2 \tilde{\chi}_{AB_iB_j}(\tau_1, \tau_2) F_i(\tau - \tau_1) F_j(\tau - \tau_2) \\ &\quad + \sum_{i,j} \int_0^\beta d\tau_1 \tilde{\chi}_{AC_{ij}}(\tau_1) F_i(\tau - \tau_1) F_j(\tau - \tau_1) + \dots, \end{aligned} \quad (3.1.75)$$

where the imaginary time-ordered response functions are defined as

$$\tilde{\chi}_{AB_i}(\tau_1) = - \left\langle T \left[\Delta \hat{B}_i(-\tau_1) \Delta \hat{A}(0) \right] \right\rangle, \quad (3.1.76)$$

$$\tilde{\chi}_{AB_iB_j}(\tau_1, \tau_2) = \frac{1}{2!} \left\langle T \left[\Delta \hat{B}_i(-\tau_1) \Delta \hat{B}_j(-\tau_2) \Delta \hat{A}(0) \right] \right\rangle, \quad (3.1.77)$$

$$\tilde{\chi}_{AC_{ij}}(\tau_1) = \frac{1}{2!} \sum_{\mathcal{P}} - \left\langle T \left[\Delta \hat{C}_{ij}(-\tau_1) \Delta \hat{A}(0) \right] \right\rangle. \quad (3.1.78)$$

The spectral representation of $\tilde{\chi}_{AB_iB_j}$ in the Matsubara frequency domain is obtained as

$$\tilde{\chi}_{AB_iB_j}(i\omega_1, i\omega_2) = \frac{1}{2!} \sum_{\mathcal{P}} \frac{A_{\lambda_1 \lambda_3} B_{2, \lambda_3 \lambda_2} B_{1, \lambda_2 \lambda_1}}{i(\omega_1 + \omega_2) + E_{\lambda_1 \lambda_3}} \left\{ \frac{P_{\lambda_1 \lambda_2}}{i\omega_1 + E_{\lambda_1 \lambda_2}} - \frac{P_{\lambda_2 \lambda_3}}{i\omega_2 + E_{\lambda_2 \lambda_3}} \right\}. \quad (3.1.79)$$

Comparing Eqs. (3.1.74) and (3.1.79), we see the similarity between the spectral representation of $\tilde{\chi}_{AB_iB_j}$ and the retarded response function $\chi_{AB_iB_j}$. By performing the analytic continuation $i\omega_i \rightarrow \omega_i + i\eta_i$, we obtain the physical response function in the real frequency domain,

$$\tilde{\chi}_{AB_iB_j}(i\omega_1, i\omega_2) \rightarrow \tilde{\chi}_{AB_iB_j}(\omega_1 + i\eta_1, \omega_2 + i\eta_2) = \chi_{AB_iB_j}(\omega_1, \omega_2). \quad (3.1.80)$$

The other physical response functions such as χ_{AB_i} and $\chi_{AC_{ij}}$ in the real frequency domain can also be obtained by analytic continuation.

Applying the above framework of analytic continuation to the current response functions in the Matsubara frequency domain, the linear and nonlinear response functions are obtained as

$$\chi^{\alpha\beta}(\omega) = \tilde{\chi}^{\alpha\beta}(\omega + i\eta), \quad (3.1.81)$$

and

$$\chi^{\alpha;\beta\gamma}(\omega_1, \omega_2) = \tilde{\chi}^{\alpha;\beta\gamma}(\omega_1 + i\eta_1, \omega_2 + i\eta_2), \quad (3.1.82)$$

respectively. When we calculate the linear and nonlinear conductivity, we replace η_i with a phenomenological scattering parameter γ for numerical convergence. Replacing $\eta_i \rightarrow \gamma$ is reasonable to obtain physically meaningful results in the finite frequency region when γ is sufficiently small [16, 111]. In the following calculations, we set the phenomenological parameters $\eta_1 = \eta_2 = \gamma$.

The current response functions can be expressed in terms of the retarded and advanced Green's functions, that are defined $\overleftrightarrow{G}^R(\mathbf{k}, z) = 1/[z - \overleftrightarrow{\mathcal{H}} + i\delta]$ and $\overleftrightarrow{G}^A(\mathbf{k}, z) = 1/[z - \overleftrightarrow{\mathcal{H}} - i\delta]$, respectively, by taking the limit $\delta \rightarrow +0$. Furthermore, we can transform the Matsubara frequency summation into the real frequency integral by using the relation $\oint_C \frac{d\omega}{2\pi i} f(\omega) A(\omega) = -\frac{1}{\beta} \sum_n A(i\omega_n)$, where $\oint_C$ represents the integral path around the poles of the Fermi distribution function $f(\omega)$ [110]. The linear response function is rewritten as

$$\begin{aligned} \chi^{\alpha\beta}(\omega) = & \frac{1}{2} \sum_{\mathbf{k}} \int_{-\infty}^{\infty} \frac{d\tilde{\omega}}{2\pi i} f(\tilde{\omega}) \text{Tr} \left[\overleftrightarrow{J}_{ab}^{\alpha\beta}(\mathbf{k}) \left(\overleftrightarrow{G}_{ba}^R(\mathbf{k}, \tilde{\omega}) - \overleftrightarrow{G}_{ba}^A(\mathbf{k}, \tilde{\omega}) \right) \right. \\ & + \overleftrightarrow{J}_{ab}^{\alpha}(\mathbf{k}) \overleftrightarrow{G}_{bi}^R(\mathbf{k}, \tilde{\omega} + \omega + i\gamma) \overleftrightarrow{\Gamma}_{ij}^{\beta}(\mathbf{k}, \omega + i\gamma) \left\{ \overleftrightarrow{G}_{ja}^R(\mathbf{k}, \tilde{\omega}) - \overleftrightarrow{G}_{ja}^A(\mathbf{k}, \tilde{\omega}) \right\} \\ & \left. + \overleftrightarrow{J}_{ab}^{\alpha}(\mathbf{k}) \left\{ \overleftrightarrow{G}_{bi}^R(\mathbf{k}, \tilde{\omega}) - \overleftrightarrow{G}_{bi}^A(\mathbf{k}, \tilde{\omega}) \right\} \overleftrightarrow{\Gamma}_{ij}^{\beta}(\mathbf{k}, \omega + i\gamma) \overleftrightarrow{G}_{ja}^A(\mathbf{k}, \tilde{\omega} - \omega - i\gamma) \right]. \end{aligned} \quad (3.1.83)$$

On the other hand, the second-order nonlinear response functions are obtained as

$$\begin{aligned} \chi^{\alpha;\beta\gamma}(\omega', \omega'') = & \sum_{\mathbf{k}} \left[\frac{1}{2} \chi^{[\alpha\beta\gamma]}(\mathbf{k}, \omega', \omega'') + \frac{1}{2} \chi^{[\alpha][\beta\gamma]}(\mathbf{k}, \omega', \omega'') + \chi^{[\alpha\beta][\gamma]}(\mathbf{k}, \omega', \omega'') \right. \\ & \left. + \chi^{[\alpha][\beta][\gamma]}(\mathbf{k}, \omega', \omega'') + (\beta \leftrightarrow \gamma, \quad \omega' \leftrightarrow \omega'') \right], \end{aligned} \quad (3.1.84)$$

where we defined

$$\chi^{[\alpha\beta\gamma]}(\mathbf{k}, \omega', \omega'') = -\frac{1}{4} \int_{-\infty}^{\infty} \frac{d\tilde{\omega}}{2\pi i} f(\tilde{\omega}) \text{Tr} \left[\overleftrightarrow{J}_{ab}^{\alpha\beta\gamma}(\mathbf{k}) \left\{ \overleftrightarrow{G}_{ba}^R(\mathbf{k}, \tilde{\omega}) - \overleftrightarrow{G}_{ba}^A(\mathbf{k}, \tilde{\omega}) \right\} \right], \quad (3.1.85)$$

$$\begin{aligned}
& \chi^{[\alpha\beta][\gamma]}(\mathbf{k}, \omega', \omega'') \\
&= -\frac{1}{4} \int_{-\infty}^{\infty} \frac{d\tilde{\omega}}{2\pi i} f(\tilde{\omega}) \text{Tr} \left[\overleftrightarrow{J}_{ab}^{\alpha\beta}(\mathbf{k}) \overleftrightarrow{G}_{bi}^R(\mathbf{k}, \tilde{\omega} + \omega'' + i\gamma) \overleftrightarrow{\Gamma}_{ij}^{\gamma}(\mathbf{k}, \omega'' + i\gamma) \overleftrightarrow{G}_{ja}^{RA}(\mathbf{k}, \tilde{\omega}) \right. \\
&\quad \left. + \overleftrightarrow{J}_{ab}^{\alpha\beta}(\mathbf{k}) \overleftrightarrow{G}_{bi}^{RA}(\mathbf{k}, \tilde{\omega}) \overleftrightarrow{\Gamma}_{ij}^{\gamma}(\mathbf{k}, \omega'' + i\gamma) \overleftrightarrow{G}_{ja}^A(\mathbf{k}, \tilde{\omega} - \omega'' - i\gamma) \right], \tag{3.1.86}
\end{aligned}$$

$$\begin{aligned}
& \chi^{[\alpha][\beta][\gamma]}(\mathbf{k}, \omega', \omega'') \\
&= -\frac{1}{4} \int_{-\infty}^{\infty} \frac{d\tilde{\omega}}{2\pi i} f(\tilde{\omega}) \text{Tr} \left[\overleftrightarrow{\Gamma}_{ab}^{\alpha}(\mathbf{k}, -\omega - 2i\gamma) \right. \\
&\quad \times \left\{ \overleftrightarrow{G}_{bi}^R(\mathbf{k}, \tilde{\omega} + \omega + 2i\gamma) \overleftrightarrow{\Gamma}_{ij}^{\beta}(\mathbf{k}, \omega' + i\gamma) \overleftrightarrow{G}_{jk}^R(\mathbf{k}, \tilde{\omega} + \omega'' + i\gamma) \overleftrightarrow{\Gamma}_{kl}^{\gamma}(\mathbf{k}, \omega'' + \gamma) \overleftrightarrow{G}_{la}^{RA}(\mathbf{k}, \tilde{\omega}) \right. \\
&\quad + \overleftrightarrow{G}_{bi}^R(\mathbf{k}, \tilde{\omega} + \omega' + i\gamma) \overleftrightarrow{\Gamma}_{ij}^{\beta}(\mathbf{k}, \omega' + i\gamma) \overleftrightarrow{G}_{jk}^{RA}(\mathbf{k}, \tilde{\omega}) \overleftrightarrow{\Gamma}_{kl}^{\gamma}(\mathbf{k}, \omega'' + i\gamma) \overleftrightarrow{G}_{la}^A(\mathbf{k}, \tilde{\omega} - \omega'' - i\gamma) \\
&\quad \left. + \overleftrightarrow{G}_{bi}^{RA}(\mathbf{k}, \tilde{\omega}) \overleftrightarrow{\Gamma}_{ij}^{\beta}(\mathbf{k}, \omega' + i\gamma) \overleftrightarrow{G}_{jk}^A(\mathbf{k}, \tilde{\omega} - \omega' - i\gamma) \overleftrightarrow{\Gamma}_{kl}^{\gamma}(\mathbf{k}, \omega'' + i\gamma) \overleftrightarrow{G}_{la}^A(\mathbf{k}, \tilde{\omega} - \omega - 2i\gamma) \right\} \Big], \tag{3.1.87}
\end{aligned}$$

$$\begin{aligned}
& \chi^{[\alpha][\beta\gamma]}(\mathbf{k}, \omega', \omega'') \\
&= -\frac{1}{4} \int_{-\infty}^{\infty} \frac{d\tilde{\omega}}{2\pi i} f(\tilde{\omega}) \text{Tr} \left[\overleftrightarrow{\Gamma}_{ab}^{\alpha}(\mathbf{k}, -\omega - 2i\gamma) \overleftrightarrow{G}_{bi}^R(\mathbf{k}, \tilde{\omega} + \omega + 2i\gamma) \overleftrightarrow{J}_{ij}^{\beta\gamma}(\mathbf{k}) \overleftrightarrow{G}_{ja}^{RA}(\mathbf{k}, \tilde{\omega}) \right. \\
&\quad \left. + \overleftrightarrow{\Gamma}_{ab}^{\alpha}(\mathbf{k}, -\omega - 2i\gamma) \overleftrightarrow{G}_{bi}^{RA}(\mathbf{k}, \tilde{\omega}) \overleftrightarrow{J}_{ij}^{\beta\gamma}(\mathbf{k}) \overleftrightarrow{G}_{ja}^A(\mathbf{k}, \tilde{\omega} - \omega - 2i\gamma) \right]. \tag{3.1.88}
\end{aligned}$$

We adopt the notation $\overleftrightarrow{G}^{RA} = \overleftrightarrow{G}^R - \overleftrightarrow{G}^A$ and $\omega = \omega' + \omega''$ for simplicity of the formulas.

Linear and nonlinear conductivity in the band representation

We discuss formulas of the linear and nonlinear conductivities in the band representation. We consider the diagonalization of the BdG Hamiltonian,

$$\left(\overleftrightarrow{U}^{\dagger}(\mathbf{k}) \right)_{ai} \left(\overleftrightarrow{\mathcal{H}}(\mathbf{k}) \right)_{ij} \left(\overleftrightarrow{U}(\mathbf{k}) \right)_{jb} = E_a(\mathbf{k}) \delta_{ab}, \tag{3.1.89}$$

where $1 \leq a, b \leq 2n$ are the indices for the energy band of Bogoliubov quasiparticles, and i, j mean the fermion's internal degrees of freedom in the Nambu space. In the following, we use the notation $\tilde{A}_{ab}$ as the band representation of $\overleftrightarrow{A}$,

$$\tilde{A}_{ab}(\mathbf{k}) = \left(\overleftrightarrow{U}^{\dagger}(\mathbf{k}) \right)_{ai} \left(\overleftrightarrow{A}(\mathbf{k}) \right)_{ij} \left(\overleftrightarrow{U}(\mathbf{k}) \right)_{jb}. \tag{3.1.90}$$

Under the same unitary transformation, the Green's functions in the band representation are given by

$$\tilde{G}_{ab}^R(\mathbf{k}, \omega) = (\omega - E_a(\mathbf{k}) + i\delta)^{-1} \delta_{ab}, \tag{3.1.91}$$

$$\tilde{G}_{ab}^A(\mathbf{k}, \omega) = (\omega - E_a(\mathbf{k}) - i\delta)^{-1} \delta_{ab}. \tag{3.1.92}$$

3.1. VERTEX CORRECTION FOR OPTICAL RESPONSES IN SUPERCONDUCTORS

Because $\overleftrightarrow{G}^R - \overleftrightarrow{G}^A$ in this representation is proportional to a delta function in the limit $\delta \rightarrow +0$, we can simplify the equations by

$$\int_{-\infty}^{\infty} \frac{d\tilde{\omega}}{2\pi i} \text{Tr} \left[\overleftrightarrow{A}_{ij}(\tilde{\omega}, \mathbf{k}) \left\{ \overleftrightarrow{G}_{ji}^R(\mathbf{k}, \tilde{\omega}) - \overleftrightarrow{G}_{ji}^A(\mathbf{k}, \tilde{\omega}) \right\} \right] = - \sum_a \tilde{A}_{aa}(\tilde{\omega} = E_a(\mathbf{k}), \mathbf{k}). \quad (3.1.93)$$

When the velocity gauge is adopted, the electric field is given by $\mathbf{E}(\omega) = i\omega \mathbf{A}(\omega)$, and the linear conductivity $\sigma^{\alpha\beta}(\omega)$ is obtained by the linear current response function,

$$\sigma^{\alpha\beta}(\omega) = \frac{\chi^{\alpha\beta}(\omega)}{i\omega}. \quad (3.1.94)$$

Using the relation (3.1.93), the linear conductivity is obtained from Eq. (3.1.83) as

$$\sigma^{\alpha\beta}(\omega) = \frac{i}{2\omega} \sum_{\mathbf{k}} \left[f_a \tilde{J}_{aa}^{\alpha\beta} + \frac{\tilde{J}_{ab}^{\alpha} \tilde{\Gamma}_{ba}^{\beta}(\omega + i\gamma) f_{ab}}{\omega - E_{ba} + i\gamma} \right], \quad (3.1.95)$$

where we define $f_a = f(E_a(\mathbf{k}))$, $f_{ab} = f_a - f_b$, and $E_{ab} = E_a(\mathbf{k}) - E_b(\mathbf{k})$ and omit the $\mathbf{k}$ description for simplicity. The second-order nonlinear optical conductivity is also rewritten by using the relation (3.1.93),

$$\begin{aligned} & \sigma^{\alpha;\beta\gamma}(\omega; \omega', \omega'') \\ &= - \frac{1}{\omega' \omega''} \chi^{\alpha;\beta\gamma}(\omega', \omega'') \\ &= - \frac{1}{4\omega' \omega''} \left[\sum_{\mathbf{k}} f_a \tilde{J}_{aa}^{\alpha\beta\gamma} + \sum_{\mathbf{k}} \frac{\tilde{\Gamma}_{ab}^{\alpha}(-\omega - 2i\gamma) \tilde{J}_{ba}^{\beta\gamma} f_{ab}}{\omega - E_{ba} + 2i\gamma} \right. \\ & \quad + \sum_{\mathbf{k}} \frac{\tilde{J}_{ab}^{\alpha\beta} \tilde{\Gamma}_{ba}^{\gamma}(\omega'' + i\gamma) f_{ab}}{\omega'' - E_{ba} + i\gamma} + \frac{\tilde{J}_{ab}^{\alpha\gamma} \tilde{\Gamma}_{ba}^{\beta}(\omega' + i\gamma) f_{ab}}{\omega' - E_{ba} + i\gamma} \\ & \quad + \sum_{\mathbf{k}} \frac{\tilde{\Gamma}_{ab}^{\alpha}(-\omega - 2i\gamma)}{\omega - E_{ba} + 2i\gamma} \left\{ \frac{\tilde{\Gamma}_{bc}^{\beta}(\omega' + i\gamma) \tilde{\Gamma}_{ca}^{\gamma}(\omega'' + i\gamma) f_{ac}}{\omega'' - E_{ca} + i\gamma} - \frac{\tilde{\Gamma}_{bc}^{\gamma}(\omega'' + i\gamma) \tilde{\Gamma}_{ca}^{\beta}(\omega' + i\gamma) f_{cb}}{\omega'' - E_{bc} + i\gamma} \right\} \\ & \quad \left. + \sum_{\mathbf{k}} \frac{\tilde{\Gamma}_{ab}^{\alpha}(-\omega - 2i\gamma)}{\omega - E_{ba} + 2i\gamma} \left\{ \frac{\tilde{\Gamma}_{bc}^{\gamma}(\omega'' + i\gamma) \tilde{\Gamma}_{ca}^{\beta}(\omega' + i\gamma) f_{ac}}{\omega' - E_{ca} + i\gamma} - \frac{\tilde{\Gamma}_{bc}^{\beta}(\omega' + i\gamma) \tilde{\Gamma}_{ca}^{\gamma}(\omega'' + i\gamma) f_{cb}}{\omega' - E_{bc} + i\gamma} \right\} \right]. \end{aligned} \quad (3.1.97)$$

After the vertex function is determined by solving the self-consistent equation, we can calculate the linear and nonlinear conductivity by using the above formulas. In the real frequency representation, the self-consistent equation for the vertex function can be expressed as a simple linear equation. We represent the vertex function as

$$\overleftrightarrow{\Gamma}_{ab}^{\alpha}(\mathbf{k}, \omega + i\gamma) = \overleftrightarrow{J}_{ab}^{\alpha}(\mathbf{k}) + \overleftrightarrow{\varphi}_{ab}(\mathbf{k}) \overleftrightarrow{\Lambda}^{\alpha}(\omega + i\gamma), \quad (3.1.98)$$

where we define

$$\overleftrightarrow{\varphi}_{ab}(\mathbf{k}) = \begin{pmatrix} \varphi_{ab}(\mathbf{k}) & 0 \\ 0 & \varphi_{ab}^{\dagger}(\mathbf{k}) \end{pmatrix}, \quad (3.1.99)$$

$$\overleftrightarrow{\Lambda}^{\alpha}(\omega + i\gamma) = \Lambda_1^{\alpha}(\omega + i\gamma) \tau_x + \Lambda_2^{\alpha}(\omega + i\gamma) \tau_y, \quad (3.1.100)$$

with the Pauli matrix in the Nambu space τ_i . When we fix the gauge so that the mean-field pair potential Δ is a real number, Λ_1^α and Λ_2^α correspond to the amplitude mode and the phase mode of collective excitations, respectively. The self-consistent equation of $\overleftrightarrow{\Lambda}^\alpha(\omega + i\gamma)$ is written as,

$$\begin{pmatrix} \Lambda_1^\alpha(\omega + i\gamma) \\ \Lambda_2^\alpha(\omega + i\gamma) \end{pmatrix} = \mathbf{M}(\omega + i\gamma) \begin{pmatrix} \Lambda_1^\alpha(\omega + i\gamma) \\ \Lambda_2^\alpha(\omega + i\gamma) \end{pmatrix} + \begin{pmatrix} L_1^J(\omega + i\gamma) \\ L_2^J(\omega + i\gamma) \end{pmatrix}. \quad (3.1.101)$$

The expressions of $\mathbf{M}(\omega + i\gamma)$ and $L_i^J(\omega + i\gamma)$ are shown in Ref. [79]. The solution of this linear equation gives the vertex function through Eq. (3.1.98).

Extension to multicomponent Cooper channels

So far we have implicitly assumed a single-component superconductor by adopting the pairing Hamiltonian, Eq. (3.1.5). It is straightforward to extend the self-consistent response approximation so that it can be applied to multicomponent superconductors. In multicomponent superconductors, multiple Cooper pairing channels allow exotic superconducting phases as chiral superconductivity [112], nematic superconductivity [113], and anapole superconductivity [114, 115].

Despite the considerable interest in exotic superconductivity, there is as yet no unambiguous confirmation of any such superconductivity. Rigorous numerical analysis of the optical response in multicomponent superconductors is required to search for strong optical probes of exotic superconductivity. We consider tow-component superconductors with two Cooper pairing channels by adopting the pairing interaction,

$$\begin{aligned} H_{\text{pair}} = & -\frac{U_1}{2V} \sum_{\mathbf{k}, \mathbf{k}'} \varphi_{ab}^{(1)}(\mathbf{k}) \varphi_{cd}^{(1)\dagger}(\mathbf{k}') c_{\mathbf{k}a}^\dagger c_{-\mathbf{k}b}^\dagger c_{-\mathbf{k}'c} c_{\mathbf{k}'d} \\ & -\frac{U_2}{2V} \sum_{\mathbf{k}, \mathbf{k}'} \varphi_{ab}^{(2)}(\mathbf{k}) \varphi_{cd}^{(2)\dagger}(\mathbf{k}') c_{\mathbf{k}a}^\dagger c_{-\mathbf{k}b}^\dagger c_{-\mathbf{k}'c} c_{\mathbf{k}'d}. \end{aligned} \quad (3.1.102)$$

The mean-field pair potential is introduced as

$$\begin{aligned} \Delta_{ab}^+(\mathbf{k}, \tau; \mathbf{A}) = & -\frac{U_1}{V} \varphi_{ab}^{(1)\dagger}(\mathbf{k}) \sum \varphi_{dc}^{(1)}(\mathbf{k}') \mathcal{F}_{cd}^+(\mathbf{k}', \tau^+, \tau^-; \mathbf{A}) \\ & -\frac{U_2}{V} \varphi_{ab}^{(2)\dagger}(\mathbf{k}) \sum \varphi_{dc}^{(2)}(\mathbf{k}') \mathcal{F}_{cd}^+(\mathbf{k}', \tau^+, \tau^-; \mathbf{A}), \end{aligned} \quad (3.1.103)$$

$$\begin{aligned} \Delta_{ab}(\mathbf{k}, \tau; \mathbf{A}) = & -\frac{U_1}{V} \varphi_{ab}^{(1)}(\mathbf{k}) \sum \varphi_{dc}^{(1)\dagger}(\mathbf{k}') \mathcal{F}_{cd}(\mathbf{k}', \tau^+, \tau^-; \mathbf{A}) \\ & -\frac{U_2}{V} \varphi_{ab}^{(2)}(\mathbf{k}) \sum \varphi_{dc}^{(2)\dagger}(\mathbf{k}') \mathcal{F}_{cd}(\mathbf{k}', \tau^+, \tau^-; \mathbf{A}), \end{aligned} \quad (3.1.104)$$

based on the Kadanoff-Baym method explained in Subsect. 3.1.2.

Let us verify the particle number conservation law. The time evolution of the total particle number is given by Eq. (3.1.32), and cancellation of the third and fourth terms in Eq. (3.1.32) is essential for the total particle number conservation law. In the presence of the multiple Cooper pairing channels, these terms are obtained as

$$\begin{aligned} & \sum \mathcal{F}_{aj}(\mathbf{k}, \tau^-, \tau^+; \mathbf{A}) \Delta_{ja}^+(\mathbf{k}, \tau; \mathbf{A}) \\ &= -\frac{U_1}{V} \left[\sum \mathcal{F}_{aj}(\mathbf{k}, \tau^-, \tau^+; \mathbf{A}) \varphi_{ja}^{(1)\dagger}(\mathbf{k}) \right] \left[\sum \varphi_{dc}^{(1)}(\mathbf{k}') \mathcal{F}_{cd}^+(\mathbf{k}', \tau^+, \tau^-; \mathbf{A}) \right] \\ & \quad - \frac{U_2}{V} \left[\sum \mathcal{F}_{aj}(\mathbf{k}, \tau^-, \tau^+; \mathbf{A}) \varphi_{ja}^{(2)\dagger}(\mathbf{k}) \right] \left[\sum \varphi_{dc}^{(2)}(\mathbf{k}') \mathcal{F}_{cd}^+(\mathbf{k}', \tau^+, \tau^-; \mathbf{A}) \right], \end{aligned} \quad (3.1.105)$$

$$\begin{aligned} & \sum \Delta_{aj}(\mathbf{k}, \tau; \mathbf{A}) \mathcal{F}_{ja}^+(\mathbf{k}, \tau^-, \tau^+; \mathbf{A}) \\ &= -\frac{U_1}{V} \left[\sum \varphi_{dc}^{(1)\dagger}(\mathbf{k}') \mathcal{F}_{cd}(\mathbf{k}', \tau^+, \tau^-; \mathbf{A}) \right] \left[\sum \varphi_{aj}^{(1)}(\mathbf{k}) \mathcal{F}_{ja}^+(\mathbf{k}, \tau^-, \tau^+; \mathbf{A}) \right] \\ & \quad - \frac{U_2}{V} \left[\sum \varphi_{dc}^{(2)\dagger}(\mathbf{k}') \mathcal{F}_{cd}(\mathbf{k}', \tau^+, \tau^-; \mathbf{A}) \right] \left[\sum \varphi_{aj}^{(2)}(\mathbf{k}) \mathcal{F}_{ja}^+(\mathbf{k}, \tau^-, \tau^+; \mathbf{A}) \right]. \end{aligned} \quad (3.1.106)$$

Therefore, we can verify that these terms cancel each other, and the total particle number conservation law is satisfied.

The formulas of the linear and second-order nonlinear conductivities are the same as Eqs. (3.1.95) and (3.1.97), respectively, while the one-photon vertex function is modified. The vertex can be represented as

$$\overleftrightarrow{\Gamma}_{ab}^\alpha(\mathbf{k}, \omega + i\gamma) = \overleftrightarrow{J}_{ab}^\alpha(\mathbf{k}) + \overleftrightarrow{\varphi}^{(1)}_{ab}(\mathbf{k}) \overleftrightarrow{\Lambda}^{(1)\alpha}(\omega + i\gamma) + \overleftrightarrow{\varphi}^{(2)}_{ab}(\mathbf{k}) \overleftrightarrow{\Lambda}^{(2)\alpha}(\omega + i\gamma), \quad (3.1.107)$$

and the self-consistent equation is written as

$$\begin{aligned} \begin{pmatrix} \mathbf{\Lambda}^{(1)\alpha}(\omega + i\gamma) \\ \mathbf{\Lambda}^{(2)\alpha}(\omega + i\gamma) \end{pmatrix} &= \begin{pmatrix} \mathbf{M}^{(11)}(\omega + i\gamma) & \mathbf{M}^{(12)}(\omega + i\gamma) \\ \mathbf{M}^{(21)}(\omega + i\gamma) & \mathbf{M}^{(22)}(\omega + i\gamma) \end{pmatrix} \begin{pmatrix} \mathbf{\Lambda}^{(1)\alpha}(\omega + i\gamma) \\ \mathbf{\Lambda}^{(2)\alpha}(\omega + i\gamma) \end{pmatrix} \\ & \quad + \begin{pmatrix} \mathbf{L}^{(1)J}(\omega + i\gamma) \\ \mathbf{L}^{(2)J}(\omega + i\gamma) \end{pmatrix}, \end{aligned} \quad (3.1.108)$$

where we use the notation $\mathbf{\Lambda}^{(i)\alpha} = (\Lambda_1^{(i)\alpha}, \Lambda_2^{(i)\alpha})^\top$ and $\mathbf{L}^{(i)J} = (L_1^{(i)J}, L_2^{(i)J})^\top$. The off-diagonal block $\mathbf{M}^{(12)}(\omega + i\gamma)$ and $\mathbf{M}^{(21)}(\omega + i\gamma)$ admix the collective modes of different Cooper channels and allow the Leggett mode and the Bardasis-Schrieffer mode [79].

Summary of the self-consistent response approximation

We have derived the formulas of linear and second-order nonlinear conductivity in the self-consistent response approximation. We applied the Kadanoff-Baym method to the equation of motion to satisfy the total particle number conservation law. Thus, the results obtained from the self-consistent response approximation are physically reasonable.

Our formulas are applicable not only to conventional superconductors but also to various unconventional superconductors, such as spin-triplet superconductors, anisotropic superconductors, and helical superconductors. The contributions of the amplitude and phase mode excitations are incorporated into the linear and nonlinear optical conductivities through the vertex correction. Furthermore, we extended the formulas to the multicomponent superconductors with multiple Cooper pairing channels. We can evaluate the contribution of the collective modes such as the Leggett mode and the Bardasis-Schrieffer mode, which can arise from multicomponent superconducting order parameters.

3.2 Setup for numerical demonstration

We study the linear and second-order nonlinear conductivities in the Rashba superconductor by the self-consistent response approximation. The effects of collective modes, which are neglected in the formulation starting from the BdG Hamiltonian, are discussed. It is shown that the amplitude mode of the s -wave superconductivity enhances the linear and nonlinear conductivities through the vertex correction. The interband pairing is essential for the enhancement. In this section, we explain the numerical demonstration setup and the characteristics of Rashba superconductors, including their multiband nature.

3.2.1 Model

The normal Hamiltonian is defined as Eqs. (2.1.1) - (2.1.6). In the following, we assume the Zeeman field as

$$\mathbf{h} = (0, h \cos \theta, h \sin \theta), \quad \theta = 60^\circ, \quad (3.2.1)$$

with θ being the angle between the magnetic field and the two-dimensional plane. We assume the magnetic field tilted from the perpendicular axis to break the C_{2z} rotation symmetry, which prohibits second-order nonlinear responses.

Next, we introduce a s -wave pairing interaction, which is not explicitly discussed in the previous chapter. In the Rashba superconductor with an in-plane component of the magnetic field, the Cooper pairs have a finite total momentum in the equilibrium superconducting state, namely helical superconducting state [76, 77]. Thus, we introduce the Cooper pairs' momentum $2\mathbf{q}$ and the s -wave pairing interaction is introduced as

$$H_{\text{pair}} = -\frac{U}{2V} \sum_{\mathbf{k}, \mathbf{k}'} (i\sigma_y)_{s_1 s_2} (-i\sigma_y)_{s_3 s_4} C_{\mathbf{k}+\mathbf{q}, s_1}^\dagger C_{-\mathbf{k}+\mathbf{q}, s_2}^\dagger C_{-\mathbf{k}'+\mathbf{q}, s_3} C_{\mathbf{k}'+\mathbf{q}, s_4}. \quad (3.2.2)$$

The mean field is obtained as

$$\Delta(\mathbf{q})_{ab} = \Delta(\mathbf{q})(i\sigma_y)_{ab}, \quad \Delta(\mathbf{q}) \in \mathbb{R}^+, \quad (3.2.3)$$

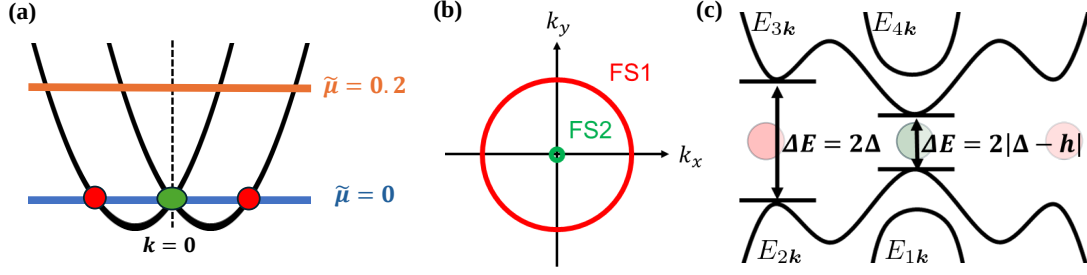


Figure 3.4: (a) Schematic parabolic bands around the Γ point (Dirac point) in the normal state. The red and green points illustrate the Fermi surfaces when the Fermi level lies on the Dirac point ($\tilde{\mu} = 0$). (b) The Fermi surfaces for $\tilde{\mu} = 0$. The FS1 exists far from the Dirac point while the FS2 is on the Dirac point. (c) Schematic illustration of the band structure in the superconducting state with the parameter $\tilde{\mu} = 0$. The energy gap is obtained as $\Delta E = 2\Delta$ around the FS1, while $\Delta E = 2|\Delta - h|$ around the FS2. From Fig. 3 of Ref. [79]. © 2025 American Physical Society

where we choose an appropriate $U(1)$ gauge.

We evaluate the condensation energy to determine the total momentum of Cooper pairs $2\mathbf{q}$. The free energy density is given by

$$\Omega(\mathbf{q}, \Delta(\mathbf{q})) = \frac{1}{2V} \sum_{\mathbf{k}, s} [\mathcal{H}_0]_{ss} + |\Delta(\mathbf{q})|^2 / 2U - \frac{T}{2V} \sum_{\mathbf{k}, n} \ln \left(1 + e^{-E_{n, \mathbf{q}}(\mathbf{k})/T} \right), \quad (3.2.4)$$

and the condensation energy is obtained as

$$F(\mathbf{q}) \equiv \Omega(\mathbf{q}, \Delta(\mathbf{q})) - \Omega(\mathbf{q}, 0). \quad (3.2.5)$$

The total momentum of Cooper pairs $2\mathbf{q}$ is determined so as to minimize the condensation energy,

$$F(\mathbf{q}) = \min_{\bar{\mathbf{q}}} F(\bar{\mathbf{q}}). \quad (3.2.6)$$

We can fix the y component of the total momentum as $q_y = 0$ because the x component of the magnetic field assume to be zero in Eq. (3.2.1). The only q_x needs to be determined by minimizing the condensation energy.

The noninteracting Hamiltonian has a Dirac point at $\mathbf{k} = \mathbf{0}$ when the magnetic field is zero. We mainly focus on the case where the Fermi level is situated at the Dirac point. The energy of the Dirac point ξ_0 is

$$\xi_0 = -4t_1 + 4t_2 = -3.2, \quad (3.2.7)$$

for our choice of the hopping parameters. We defined $\tilde{\mu}$ as the chemical potential from the Dirac point, $\tilde{\mu} \equiv \mu - \xi_0$. When we set the chemical potential as $\tilde{\mu} = 0$, the Fermi level lies

on the Dirac point under zero magnetic field [see Fig. 3.4(a)]. In this case, one of the two spin-split Fermi surfaces, ‘Fermi surface 1’ (FS1), exists far from the Dirac point, and the other ‘Fermi surface 2’ (FS2) is the point at the Dirac point [Fig. 3.4(b)]. In the following part, we assume $\tilde{\mu} = 0.0$ unless otherwise mentioned.

The numerical calculations are conducted within the N^2 -discretized Brillouin zone. Unless stated otherwise, N is set to 1600. For numerical convergence, we introduce a finite temperature $T = 10^{-4}$ for the Fermi-Dirac distribution function. We evaluate the linear and nonlinear conductivities by using the self-consistent response approximation and discuss the effects of the collective modes.

3.2.2 Intraband pairing and interband pairing

Before showing the numerical results, we elucidate the intraband and interband pairing in the model. In Secs. 3.3 and 3.4, we will show that interband pairing plays an essential role in enhancing optical responses via collective modes. The superconducting fitness can capture intraband pairing and interband pairing [116, 117]. We define the intraband superconducting fitness F_A and the interband superconducting fitness F_C at momentum $\mathbf{k}$ by

$$F_A(\mathbf{k})(i\sigma_y) = \mathcal{H}'_0(\mathbf{k})[\Delta(i\sigma_y)] + [\Delta(i\sigma_y)]\mathcal{H}'_0{}^\top(-\mathbf{k}), \quad (3.2.8)$$

$$F_C(\mathbf{k})(i\sigma_y) = \mathcal{H}'_0(\mathbf{k})[\Delta(i\sigma_y)] - [\Delta(i\sigma_y)]\mathcal{H}'_0{}^\top(-\mathbf{k}). \quad (3.2.9)$$

We define $\mathcal{H}'_0(\mathbf{k}) \equiv \mathbf{c}(\mathbf{k}) \cdot \boldsymbol{\sigma} / |\mathbf{c}(\mathbf{k})|$ for the noninteracting Hamiltonian with the form $\mathcal{H}_0(\mathbf{k}) = c_0(\mathbf{k}) + \mathbf{c}(\mathbf{k}) \cdot \boldsymbol{\sigma}$. In our model, $\mathcal{H}'_0(\mathbf{k})$ is obtained as $\mathcal{H}'_0(\mathbf{k}) = [\mathcal{H}_0(\mathbf{k}) - \xi(\mathbf{k})] / |\mathbf{g}(\mathbf{k}) + \mathbf{h}|$. When the Cooper pairs’ total momentum $2\mathbf{q}$ is sufficiently small and ignorable, the intraband and interband pairing is characterized by the fitness as

$$\text{Tr}[F_A(\mathbf{k})^\dagger F_A(\mathbf{k})] = 8\Delta^2 g(\mathbf{k})^2 / |\mathbf{g}(\mathbf{k}) + \mathbf{h}|^2, \quad (3.2.10)$$

$$\text{Tr}[F_C(\mathbf{k})^\dagger F_C(\mathbf{k})] = 8\Delta^2 h^2 / |\mathbf{g}(\mathbf{k}) + \mathbf{h}|^2. \quad (3.2.11)$$

These formulas indicate that the interband pairing is dominant around the Dirac point because the Rashba spin-orbit coupling vanishes at the Dirac point. On the other hand, the intraband pairing is dominant around the FS1 as usual. The energy gap also differs between the k -space region around the FS1 and FS2 [see Fig. 3.4(c)]. The energy gap on the FS1 is obtained as $\Delta E = 2\Delta$, while the gap at the FS2 is estimated as $\Delta E = 2|\Delta - h|$ [57]. Thus, the superconducting gap around the FS1 is robust against the Zeeman field although it closes on the FS2 at the topological transition.

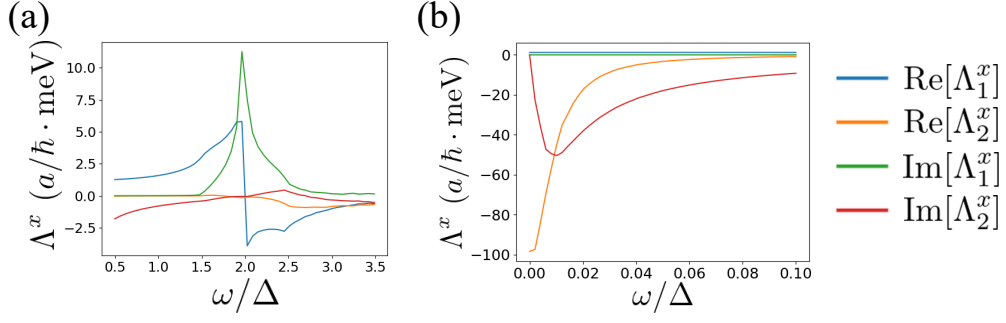


Figure 3.5: Frequency dependence of Λ_i^x [Eq. (3.1.100)] in the vertex function, Eq. (3.1.98). Λ_1^x and Λ_2^x originate from the amplitude and phase modes, respectively. We set the parameters $\tilde{\mu} = 0.0$, $U = 1.15$, $h = 0.02$, and $\gamma = 8 \times 10^{-4}$. Results in (a) the moderate frequency region $0.5 \leq \Omega/\Delta \leq 3.5$ and (b) the low-frequency region $0 \leq \Omega/\Delta \leq 0.1$. From Fig. 4 of Ref. [79]. © 2025 American Physical Society

3.2.3 Vertex function

In this subsection, we show the self-consistent solution of the vertex function and discuss its characteristic properties. By comparing Eqs. (3.1.51), (3.1.98), and (3.1.100), we can verify the correspondence between Λ_i^x and the collective mode excitations. Figure 3.5 shows the frequency dependence of $\Lambda_i^x(\omega + i\gamma)$. The peak of $\text{Im}[\Lambda_1^x]$ around $\omega = 2\Delta$ originates from the amplitude mode known as the Higgs mode. The nearly ω^{-1} divergence of $\text{Im}[\Lambda_2^x]$ in the low-frequency region $\omega \ll 2\Delta$ may be attributed to the phase mode known as the Nambu-Goldstone mode. The behavior of Λ_2^x in the low-frequency region shown in Fig. 3.5(b) can be described as

$$\Lambda_2^x(\omega + i\gamma) \propto \frac{i}{\omega + i\gamma}, \quad (3.2.12)$$

by introducing the phenomenological scattering rate γ to cut off the divergence.

3.3 Linear optical responses

In this section, we show the linear optical conductivity with and without vertex correction and discuss the roles of the collective modes. In the case of $\tilde{\mu} = 0.0$, where the Fermi level lies on the Dirac point, the interband pairing is dominant around the Dirac point. We will show that the amplitude mode enhances the longitudinal linear conductivity in this case. For s -wave superconductors, the enhancement by the amplitude mode is unique to the multiband system. We verify that interband pairing is essential for the enhancement of linear conductivity by the amplitude mode.

In the following, the linear conductivity obtained with (without) vertex correction is

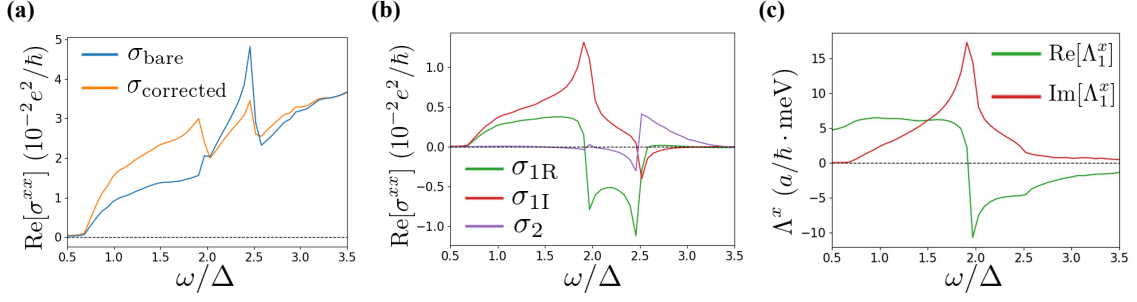


Figure 3.6: Frequency dependence of (a) the linear conductivity $\text{Re}[\sigma^{xx}]$, (b) its components defined by Eq. (3.3.1), and (c) the vertex function Λ_1^x . (a) The bare and corrected linear conductivities are compared to clarify the role of vertex correction. (b) The contribution of the vertex correction arising from the amplitude (Higgs) mode and the phase (NG) mode to the linear conductivity. The amplitude mode gives σ_{1R} and σ_{1I} , while the phase mode gives σ_2 . We set the parameters as $\tilde{\mu} = 0.0$ and $h = 0.05$. From Fig. 5 of Ref. [79]. © 2025 American Physical Society

referred to as the corrected (bare) conductivity. The main result of this section is presented in Fig. 3.6(a), which shows the bare and corrected linear conductivities $\text{Re}[\sigma^{xx}]$. We see that the linear conductivity is enhanced by the vertex correction in the low frequency region $\omega/\Delta \leq 2.0$. To identify the causes of enhancement, we decompose the contribution of the vertex by using Eqs. (3.1.95), (3.1.98), and (3.1.100). The correction to the linear conductivity can be written as

$$\sigma_{\text{corrected}} - \sigma_{\text{bare}} = \sigma_1 + \sigma_2, \quad (3.3.1)$$

with $\sigma_1 = \sigma_{1R} + \sigma_{1I}$, where σ_{1R} , σ_{1I} , and σ_2 are contributions proportional to $\text{Re}[\Lambda_1^x]$, $\text{Im}[\Lambda_1^x]$, and Λ_2^x , respectively. Figure 3.3.1(b) shows that Λ_1^x makes the dominant contribution to the enhancement of the linear conductivity. Because the vertex Λ_1^x (Λ_2^x) comes from the amplitude (phase) mode, we conclude that the amplitude mode enhances the linear conductivity.

3.3.1 Multiband effect

To discuss the multiband effects on the linear conductivity, we then compare our Rashba superconductor model with the single-band s -wave superconductor model. In Ref. [81], it was shown that the linear conductivity $\text{Re}[\sigma^{xx}]$ is suppressed by the vertex correction in the single-band s -wave superconductor under supercurrent. We reproduce the suppression of the corrected linear conductivity in Fig. 3.7. We find that the vertex Λ_2^x owing to the phase gives a destructive contribution, while the effect of the amplitude mode through Λ_1^x is negligible in this case.

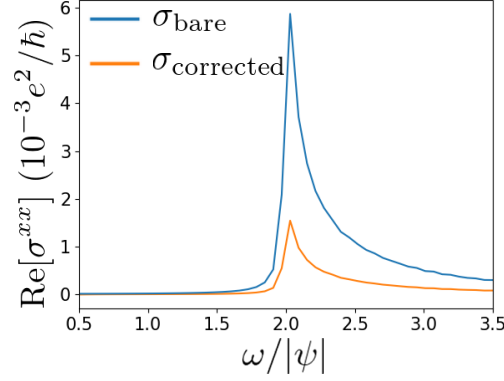


Figure 3.7: The linear conductivity $\text{Re}[\sigma^{xx}]$ in the single-band model. The numerical result reproduces the suppression of the corrected linear conductivity in Ref. [81]. From Fig. 6 of Ref. [79]. © 2025 American Physical Society

We can analytically show that the contribution of the amplitude mode is negligible in the single-band superconductor. The BdG Hamiltonian for single-band s -wave superconductors is given by

$$\mathcal{H}(\mathbf{k}) = \begin{pmatrix} \xi_1(\mathbf{k}) & \psi \\ \psi & -\xi_2(\mathbf{k}) \end{pmatrix}, \quad \psi \in \mathbb{R}^+, \quad (3.3.2)$$

where we choose an appropriate $U(1)$ gauge. By introducing the electric current operator and the one-photon vertex function,

$$J^\alpha(\mathbf{k}) = J_0(\mathbf{k}) + J_3(\mathbf{k})\sigma_3, \quad (3.3.3)$$

$$\Gamma^\alpha(\mathbf{k}, \omega + i\gamma) = J^\alpha(\mathbf{k}) + \Lambda_1^\alpha(\omega + i\gamma)\sigma_1 + \Lambda_2^\alpha(\omega + i\gamma)\sigma_2, \quad (3.3.4)$$

the resonant component of the linear optical conductivity is obtained as

$$\sigma^{\alpha\beta}(\omega) = \sum_{\mathbf{k}} \frac{\pi\psi J_3^\alpha}{2\Delta E^2} \left\{ \frac{-\bar{\xi}\Lambda_1^\beta + J_3^\beta\psi}{\Delta E} + i\Lambda_2^\beta \right\} \delta(\omega - 2\Delta E), \quad (3.3.5)$$

where $\bar{\xi}(\mathbf{k})$ is defined as $\bar{\xi}(\mathbf{k}) = (\xi_1(\mathbf{k}) + \xi_2(\mathbf{k}))/2$. The interband energy difference is given by $2\Delta E = 2\sqrt{\psi^2 + \bar{\xi}(\mathbf{k})^2}$, and the minimum energy gap is 2ψ . The resonant component is enhanced around $\omega \sim 2\psi$ because of the prefactor ΔE^{-2} and the peak of the joint density of states. However, the contribution of the amplitude mode through Λ_1^β is suppressed there by the prefactor $\bar{\xi} \sim 0$. Moreover, the vertex arising from the amplitude mode Λ_1^β is relatively small compared to Λ_1^α originates from the weighted integral of $\bar{\xi}(\mathbf{k})$ around the Fermi surface [87]. Therefore, the effect of the amplitude mode is negligible in the single-band superconductors.

In contrast, in the multiband systems, the amplitude mode can enhance the optical conductivity through the vertex correction Λ_1^β . Figure 3.6(b) shows the enhancement by

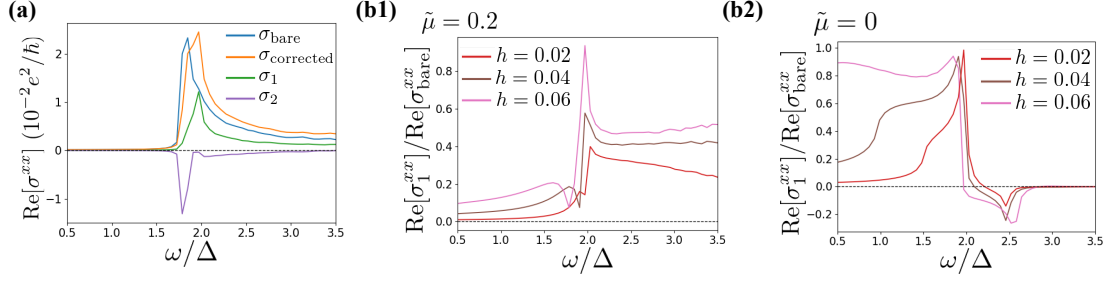


Figure 3.8: (a) The linear conductivity $\text{Re}[\sigma^{xx}]$ for the effective chemical potential $\tilde{\mu} = 0.2$ and the magnetic field $h = 0.06$. (b1) and (b2) The ratio of $\text{Re}[\sigma_1^{xx}]$ to $\text{Re}[\sigma_{\text{bare}}^{xx}]$ for various magnitude of the magnetic field. We set $\tilde{\mu} = 0.2$ and $\tilde{\mu} = 0$ in (b1) and (b2), respectively. From Fig. 7 of Ref. [79]. © 2025 American Physical Society

the collective mode in the Rashba superconductors. In our model, the Rashba spin-orbit coupling and the Zeeman field split the energy bands and allow the contributions by the amplitude mode, which is prohibited in the single-band model. The multiband effect is essential to enhance the linear conductivity by the amplitude mode in *s*-wave superconductors.

Interband pairing

To further clarify the multiband effects, we focus on the role of interband pairing. The resonant component of the enhanced linear conductivity in the region $\omega/\Delta < 2.0$ originates from momentum space around the Dirac point [see Figs. 3.4(a) and 3.4(c)]. Given Eqs. (3.2.10) and (3.2.11), the interband superconducting fitness is much larger than the intraband superconducting fitness around the Dirac point. In contrast, the interband pairing is absent in single-band *s*-wave superconductors. Therefore, the interband pairing is expected to play an essential role in the enhancement of linear conductivity shown in Fig. 3.6(a).

We then show the numerical results that indicate the importance of the interband pairing for the enhancement by the amplitude mode. Here, we consider the $\tilde{\mu} = 0.2$ case, where the Fermi surface is away from the Dirac point [Fig. 3(a)]. Although intraband pairing is dominant in this case, we can increase the interband pairing by increasing the magnetic field. Thus, we expect the vertex correction arising from the amplitude mode to increase with increasing magnetic field. Figure 3.8(a) shows that the contribution due to the amplitude mode, $\sigma_1 = \sigma_{1R} + \sigma_{1I}$, is cooperative to the bare conductivity σ_{bare} . When increasing the magnitude of the magnetic field, the ratio $\text{Re}[\sigma_1]/\text{Re}[\sigma_{\text{bare}}]$ increases [Fig. 3.8(b1)]. It is consistent with our expectations. A sharp peak caused by the frequency dependence of Λ_1^x [Fig. 3.9(b1)] appears in the ratio $\text{Re}[\sigma_1]/\text{Re}[\sigma_{\text{bare}}]$, and the peak develops with increasing the magnetic field. This indicates that the interband pairing is required

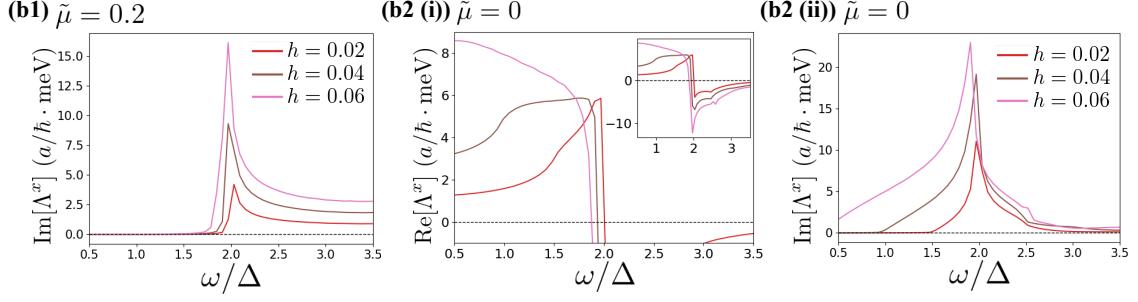


Figure 3.9: Frequency dependence of $\Lambda_1^x(\omega + i\gamma)$ in the vertex function [Eq. (3.1.100)]. Λ_1^x originates from the amplitude mode. The parameters in panels (b1) and (b2) are the same as Figs. 3.8(b1) and 3.8(b2), respectively. (b1) and (b2 (ii)): The imaginary part of Λ_1^x . (b2 (i)): The real part of Λ_1^x . From Fig. 8 of Ref. [79]. © 2025 American Physical Society

for the enhancement of the linear conductivity by the amplitude mode.

Similarly, in the case of $\tilde{\mu} = 0$, the ratio $\text{Re}[\sigma_1]/\text{Re}[\sigma_{\text{bare}}]$ in Fig. 3.8(b2) shows the characteristic frequency dependence caused by Λ_1^x [Figs. 3.9(b2 (i)) and 3.9(b2 (ii))]. At a small magnetic field $h = 0.02$, the peak of the ratio around $\omega/\Delta \simeq 2.0$ appears even more clearly than that for $\tilde{\mu} = 0.2$. This is consistent with that the interband pairing is significant around the Dirac point even for a small magnetic field and our conclusion that the interband pairing allows the enhancement of optical responses by the amplitude mode.

In contrast to the low-frequency region discussed above, the vertex correction suppresses the linear conductivity in the high-frequency region $\omega/\Delta > 2.0$, as shown in Fig. 3.6(a). The suppression is also the result of the amplitude mode [see Fig. 3.6(b)]. The sign of the vertex $\text{Re}[\Lambda_1]$ changes around $\omega/\Delta \sim 2.0$ [Fig. 3.6(c)], and σ_{1R} becomes negative. This effect of the amplitude mode does not appear in the Rashba model for $\tilde{\mu} = 0.2$ and in the single-band model. Therefore, the vertex correction that suppresses the linear conductivity is also a characteristic property of the Dirac point.

We summarize the results of the linear conductivity in s -wave Rashba superconductors. We have shown that the amplitude mode of the order parameter enhances the linear conductivity $\text{Re}[\sigma^{xx}]$ in the low-frequency region $\omega/\Delta < 2.0$. This enhancement in clean s -wave superconductors is unique to multiband superconductors. In particular, the interband pairing is essential for the enhancement. Therefore, the linear conductivity is significantly enhanced in the model with $\tilde{\mu} = 0.0$, where interband pairing is dominant around the Dirac point. The peculiar quantum-geometric properties of Dirac electrons are also important for the large optical conductivity. When the Fermi level lies on the Dirac point, the linear conductivity is expected to be large since it is related to the quantum-geometric quantities. Indeed, this is the reason why the linear conductivity for $\tilde{\mu} = 0.0$ [Fig. 3.6(a)] is larger than that for $\tilde{\mu} = 0.2$ [Fig. 3.6(a)]. Because the vertex correction from the collective modes

is expected to be of the same order as the bare conductivity, the vertex correction term σ_1 is also enhanced cooperatively by the interband pairing and the quantum-geometric properties both of which are enhanced around the Dirac point.

3.4 Nonlinear optical responses

In this section, we show the results of the nonlinear optical conductivity. In particular, we focus on the photocurrent conductivity for which a theoretical calculation without vertex correction predicted the sign reversal at the topological transition of superconducting states [57]. We show that the sign reversal occurs robustly when collective-mode excitations are included. Furthermore, the resonant components showing the sign reversal in the low-frequency region are enhanced by the amplitude mode. The sign reversal originates from the band inversion, which is directly related to the topological transition, and therefore it can be used as an indicator of topological superconductivity.

3.4.1 Calculation method of photocurrent conductivity

The photocurrent conductivity is defined as

$$\sigma_{\text{PC}}^{\alpha;\beta\gamma}(\omega) \equiv \lim_{\delta\omega \rightarrow 0} \sigma^{\alpha;\beta\gamma}(2\delta\omega; \omega + \delta\omega, -\omega + \delta\omega), \quad (3.4.1)$$

where ω is a frequency of an injection monochromatic light. Although the second-order nonlinear conductivity $\sigma^{\alpha;\beta\gamma}$ has been formulated in the previous sections, we need to take care of the numerical calculation because the calculation with the vertex correction can be numerically unstable when the massless collective mode emerges. The formula of the photocurrent conductivity, Eq. (3.1.97), has terms containing the vertex function $\tilde{\Gamma}_{ab}^{\alpha}(-2\delta\omega - 2i\gamma)$ such as

$$\lim_{\delta\omega \rightarrow 0} \frac{\tilde{\Gamma}_{ab}^{\alpha}(-2\delta\omega - 2i\gamma) \tilde{J}_{ba}^{\beta\gamma} f_{ab}}{4(\omega + \delta\omega)(\omega - \delta\omega)(\omega - E_{ba} + 2i\gamma)}. \quad (3.4.2)$$

Because a part of the vertex function $\Lambda_2^{\alpha}(\delta\omega + i\gamma)$ diverges in the limit $\delta\omega \rightarrow 0$ and $\gamma \rightarrow 0$ (clean limit), we should examine possible divergence of the photocurrent conductivity. However, the divergent vertex function Λ_2^{α} does not necessarily give the divergent photocurrent conductivity. For example, in the single-band s -wave model, the contribution of Λ_2^{α} to Eq. (3.4.2) has a prefactor $\delta\omega$ [81]. Given $\Lambda_2^{\alpha} \delta\omega^{-1}$ in the limit $\delta\omega \rightarrow 0$ and $\gamma \rightarrow 0$, the term proportional to $\delta\omega \Lambda_2^{\alpha}$ gives a finite contribution to the photocurrent conductivity without divergence.

To avoid potential difficulty caused by the divergent vertex function, we compare two methods for taking the clean and low different-frequency limit, $\delta\omega \rightarrow 0$ and $\gamma \rightarrow 0$. One is

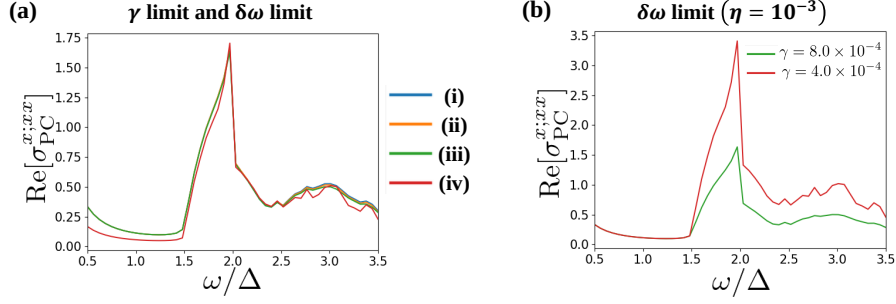


Figure 3.10: (a) Comparison of the photocurrent conductivity $\text{Re}[\sigma_{\text{PC}}^{x;xx}(\omega)]$ obtained in (i) the $\delta\omega$ -limit and (ii)-(iv) the γ -limit. (i) $\delta\omega$ -limit with $\gamma = 8.0 \times 10^{-4}$. (ii) γ -limit with $\eta = 10^{-4}$ and $\gamma = 8.0 \times 10^{-4}$. (iii) γ -limit with $\eta = 10^{-3}$ and $\gamma = 8.0 \times 10^{-4}$. (iv) $\text{Re}[\sigma_{\text{PC}}^{x;xx}(\omega)]/2$ in γ -limit with $\eta = 10^{-3}$ and $\gamma = 4.0 \times 10^{-4}$. The result is divided by 2 to show the γ^{-1} behavior of $\text{Re}[\sigma_{\text{PC}}^{x;xx}(\omega)]$. (b) $\text{Re}[\sigma_{\text{PC}}^{x;xx}(\omega)]$ obtained in the $\delta\omega$ -limit with $\gamma = 8.0 \times 10^{-4}$ and $\gamma = 4.0 \times 10^{-4}$. The resonant component appears at $\omega/\Delta > 1.5$ and is proportional to γ^{-1} , indicating that the magnetic injection current is dominant. The parameters of the model are set to $\tilde{\mu} = 0.0$, $U = 1.15$, and $h = 0.02$. The unit of the vertical axis is $(e^3/\hbar \cdot a/\text{meV})$. From Fig. 9 of Ref. [79]. © 2025 American Physical Society

the $\delta\omega$ -limit, where we assume a finite γ and take the limit $\delta\omega \rightarrow 0$. The vertex function is regularized as

$$\Gamma^\alpha(-2\delta\omega - 2i\gamma) \rightarrow \Gamma^\alpha(-2i\gamma). \quad (3.4.3)$$

The other is the γ -limit, where we first take the clean limit $\gamma \rightarrow 0$. The frequency dependence of the vertex function, plotted in Fig. 3.5(b), can be numerically fitted by using a fitting function,

$$\Gamma^\alpha(\omega + i\gamma) \simeq \frac{iA}{\omega + iB}, \quad (3.4.4)$$

with fitting parameters A and B . The obtained value of B is almost equal to γ . Therefore, for a sufficiently small frequency difference $\delta\omega$ and the limit $\gamma \rightarrow 0$, we adopt the asymptotic form,

$$\Gamma^\alpha(-2\delta\omega - 2i\gamma) \rightarrow \frac{iA}{-2\delta\omega}. \quad (3.4.5)$$

In numerical calculation, we introduce a sufficiently small parameter η and the frequency difference $2\delta\omega$ is set as $2\delta\omega = \eta\omega$. The numerical results are physically meaningful if the two methods give the same results. Figure 3.10(a) compares $\text{Re}[\sigma_{\text{PC}}^{x;xx}(\omega)]$ obtained in the γ -limit and the $\delta\omega$ -limit. It is shown that the numerical results are almost equivalent between the two methods and do not depend on the choice of η in the $\delta\omega$ -limit. Thus, we

can obtain physically meaningful results, which are independent of the treatment of the massless collective modes.

Next, we discuss the dependence of the photocurrent conductivity on the phenomenological scattering rate γ to verify that the intrinsic effect is dominant in our results. Figure 3.10(b) shows $\text{Re}[\sigma_{\text{PC}}^{x;xx}(\omega)]$ obtained in the $\delta\omega$ -limit with different γ . We see that the magnitude of the resonant contribution is doubled when γ is decreased to half. Thus, the photocurrent conductivity shows the γ^{-1} dependence signifying the dominance of the magnetic injection current, which is one of the intrinsic mechanisms of photocurrent conductivity [20, 118–120]. From these analyses, it is verified that our formulation can adequately describe the intrinsic contribution of the photocurrent conductivity in the Rashba superconductors.

3.4.2 Sing reversal of photocurrent conductivity

We carry out numerical calculations of the photocurrent conductivity around the topological transition. The results with and without the vertex correction are presented. We show that the sign reversal is robust against the contribution of collective modes.

When we set the parameters as $\tilde{\mu} = 0.0$ and $U = 1.15$, the model describes topologically trivial superconducting states in the low magnetic field region $0 \leq h \leq 0.06$ but becomes topological superconducting states in the high magnetic field region $0.07 \leq h \leq 0.09$. Figures 3.11(a1) and 3.11(b1) plot the bare photocurrent conductivity, while the corrected one with vertex correction is shown in Figs. 3.11(a2) and 3.11(b2). Qualitative behaviors, such as the shift in the frequency dependence of the photocurrent, are similar between the bare and corrected photocurrent conductivities. The shift reflects the suppression of the energy gap under the magnetic field. The energy gap in the trivial state becomes smaller with h being closer to the critical value h_c and closes at the topological transition. After the topological transition, the energy gap reopens as the magnetic field is increased. Following changes of the energy gap, the resonant contribution of the photocurrent conductivity $\text{Re}[\sigma_{\text{PC}}^{x;xx}]$ shows the frequency shift.

In Sec. 2, we demonstrated the sign reversal of the photocurrent conductivity in the low-frequency region $\omega/\Delta < 2.0$ without the vertex correction. The sign reversal of the bare photocurrent conductivity is reproduced in Fig. 3.11(a1) and 3.11(b1). The sign reversal behavior appears even when we take into account the vertex correction. As shown in Figs. 3.11(a2) and 3.11(b2), the sign of the corrected photocurrent conductivity also reverses at the topological transition. Therefore, the sign reversal is robust against the contributions of collective modes, and thus this phenomenon is expected to be useful as an indicator of topological superconductivity.

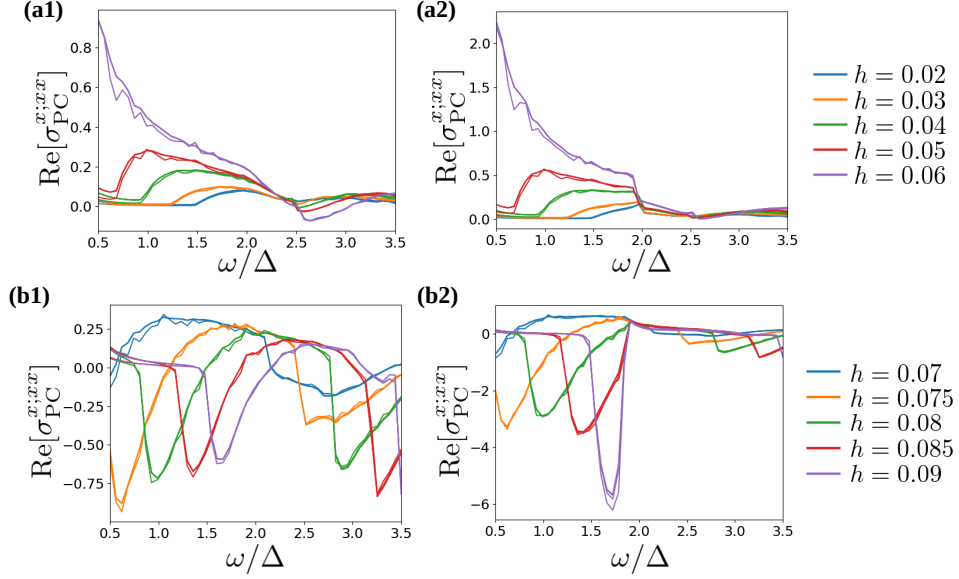


Figure 3.11: The photocurrent conductivity $\text{Re}[\sigma_{\text{PC}}^{x;xx}]$ in the topologically trivial states [(a1), (a2)] and the topological superconducting states [(b1), (b2)] with varying the magnitude of the magnetic field h . (a1) and (b1) show the bare photocurrent conductivity without vertex correction, while (a2) and (b2) plot the corrected conductivity with vertex correction. For each magnetic field, the calculations are performed in the γ -limit and the ω -limit methods with setting the same parameter sets η and γ as in Fig. 3.10(a). We rescale the results with respect to γ as we have done in Fig. 3.10(a) and plot the results with the same color for each h . The parameters are set as $\tilde{\mu} = 0.0$ and $U = 1.15$. From Fig. 10 of Ref. [79]. © 2025 American Physical Society

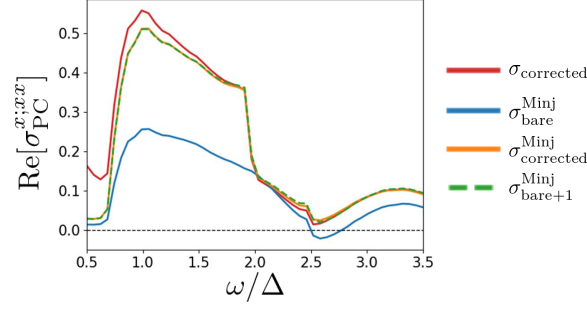


Figure 3.12: The photocurrent conductivity $\text{Re}[\sigma_{\text{PC}}^{x;xx}]$ and its components. The total photocurrent conductivity $\sigma_{\text{corrected}}$ with vertex correction (red line) is compared with the bare and corrected magnetic injection currents $\sigma_{\text{bare}}^{\text{Minj}}$ (blue line) and $\sigma_{\text{corrected}}^{\text{Minj}}$ (orange line). We also show $\sigma_{\text{bare}+1}^{\text{Minj}}$ (green dashed line), which is the magnetic injection current obtained by taking into account only the vertex correction as a result of the amplitude mode. The parameters are set as $\tilde{\mu} = 0.0$, $h = 0.05$ and $U = 1.15$. The unit of the vertical axis is $(e^3/\hbar \cdot a/\text{meV})$. From Fig. 11 of Ref. [79]. © 2025 American Physical Society

3.4.3 Magnetic injection current

For a step to clarify the origin of the sign reversal phenomenon, we discuss the magnetic injection current. The bare magnetic injection current, which is calculated without vertex corrections, is formulated as

$$\sigma_{\text{bare}}^{\text{Minj}} = -\frac{\pi}{4\gamma} \sum_{a,b,\mathbf{k}} \Delta \tilde{J}_{ab}^x \frac{\text{Re}[\tilde{J}_{ba}^x \tilde{J}_{ab}^x]}{E_{ba}^2} F_{ab}, \quad (3.4.6)$$

where we define $F_{ab} = f_{ab} \delta(\omega - E_{ba})$ and $\Delta \tilde{J}_{ab}^\alpha = \tilde{J}_{aa}^\alpha - \tilde{J}_{bb}^\alpha$.

To discuss the magnetic injection current in the self-consistent approximation, we introduce the corrected magnetic injection current as

$$\sigma_{\text{corrected}}^{\text{Minj}} = -\frac{\pi}{4\gamma} \sum_{a,b,\mathbf{k}} \Delta \tilde{\Gamma}_{ab}^{x2A}(0) \frac{\text{Re}[\tilde{\Gamma}_{ba}^{xR}(\omega) \tilde{\Gamma}_{ab}^{xR}(-\omega)]}{E_{ba}^2} F_{ab}, \quad (3.4.7)$$

where we use the following notations

$$\Delta \tilde{\Gamma}_{ab}^{x2A}(\omega) = \tilde{\Gamma}_{aa}^x(\omega - 2i\gamma) - \tilde{\Gamma}_{bb}^x(\omega - 2i\gamma), \quad (3.4.8)$$

$$\tilde{\Gamma}_{ab}^{xR}(\omega) = \tilde{\Gamma}_{ab}^x(\omega + i\gamma). \quad (3.4.9)$$

This contribution is obtained from the formula for the second-order nonlinear responses [Eq. (3.1.97)]. In the formula for the corrected magnetic injection current, the electric current operator appearing in the bare magnetic injection current is replaced with the one-photon vertex $\tilde{\Gamma}$, which includes the effects of the collective mode. Figure 3.12 plots the total corrected photocurrent conductivity $\sigma_{\text{corrected}}$ and the component $\sigma_{\text{corrected}}^{\text{Minj}}$. We

see that $\sigma_{\text{corrected}}^{\text{Minj}}$ gives a dominant contributions to the photocurrent conductivity in this frequency region. This is consistent with our expectation, and thus the magnetic injection current is dominant even in the self-consistent approximation, which takes into account the vertex correction.

Enhancement by the amplitude mode

Comparing the bare and corrected magnetic injection current in Fig. 3.12, we find that the magnetic injection current is enhanced by the vertex correction. Let us verify that the amplitude mode is essential for the enhancement. To elucidate the importance of the amplitude mode, we calculate $\sigma_{\text{bare}+1}^{\text{Minj}}$ where the one-photon vertex in $\sigma_{\text{bare}+1}^{\text{Minj}}$ is evaluated by taking into account only the bare electric current operator and Λ_1^x . The amplitude mode influences $\sigma_{\text{bare}+1}^{\text{Minj}}$ through the vertex Λ_1^x , while the effect of the phase mode is ignored. Figure 3.12 shows that $\sigma_{\text{bare}+1}^{\text{Minj}}$ is almost equivalent to $\sigma_{\text{corrected}}^{\text{Minj}}$. Therefore, we conclude that the enhancement of the photocurrent conductivity originates from the amplitude mode, whereas the phase mode is ignorable.

3.4.4 Mechanism of the sign reversal

We discuss the mechanism of sign reversal of the photocurrent conductivity associated with the topological transition. The resonant photocurrent conductivity in the low-frequency region $\omega/\Delta < 2.0$ originates from the low-energy quasiparticle excitation [57], whose energies of $E_{2\mathbf{k}}$ and $E_{3\mathbf{k}}$ in our model. Note that we have defined the Bogoliubov quasiparticle's bands as $E_{1\mathbf{k}} \leq E_{2\mathbf{k}} \leq 0 \leq E_{3\mathbf{k}} \leq E_{4\mathbf{k}}$ [see Figure 3.4(c)]. Therefore, we focus on the interband transition between the $E_{2\mathbf{k}}$ and $E_{3\mathbf{k}}$ bands, namely B2 transition in Subsect. 2.2.2. In the zero temperature limit, the magnetic injection current originating from the B2 transition $\sigma_{\text{corrected,B2}}^{\text{Minj}}$ is given by

$$\sigma_{\text{corrected,B2}}^{\text{Minj}}(\omega) = -\frac{\pi}{4\gamma} \sum_{\mathbf{k}} \Delta \tilde{\Gamma}_{23}^{x2A}(0) \frac{\text{Re}[\tilde{\Gamma}_{32}^{xR}(\omega) \tilde{\Gamma}_{23}^{xR}(-\omega)]}{E_{32}^2} \delta(\omega - E_{32}). \quad (3.4.10)$$

We can show that the factor $\text{Re}[\tilde{\Gamma}_{32}^{xR}(\omega) \tilde{\Gamma}_{23}^{xR}(-\omega)]$ must be positive [79]. Therefore, the sign of $\sigma_{\text{corrected,B2}}^{\text{Minj}}$ is determined by the difference in the one-photon vertex $\Delta \tilde{\Gamma}_{23}^{x2A}(0)$. Because the resonant component of $\text{Re}[\sigma_{\text{PC}}^{x;xx}]$ in the low-frequency region arises from the momentum space around $\mathbf{k} = \mathbf{0}$, the sign reversal of $\text{Re}[\sigma_{\text{PC}}^{x;xx}]$ is caused by $\Delta \tilde{\Gamma}_{23}^{x2A}(0)$ around $\mathbf{k} = \mathbf{0}$, which is shown in Fig. 3.13. We find that the sign of $\Delta \tilde{\Gamma}_{23}^{x2A}(0)$ opposite between the trivial state [Fig. 3.13(a)] and the topological state [Fig. 3.13(b)] around $\mathbf{k} = \mathbf{0}$.

We can explain the reason why the sign of $\Delta \tilde{\Gamma}_{23}^{x2A}(0)$ changes at the topological transition, with the similar discussion to Sec. 2.3. The sign change is associated with the band

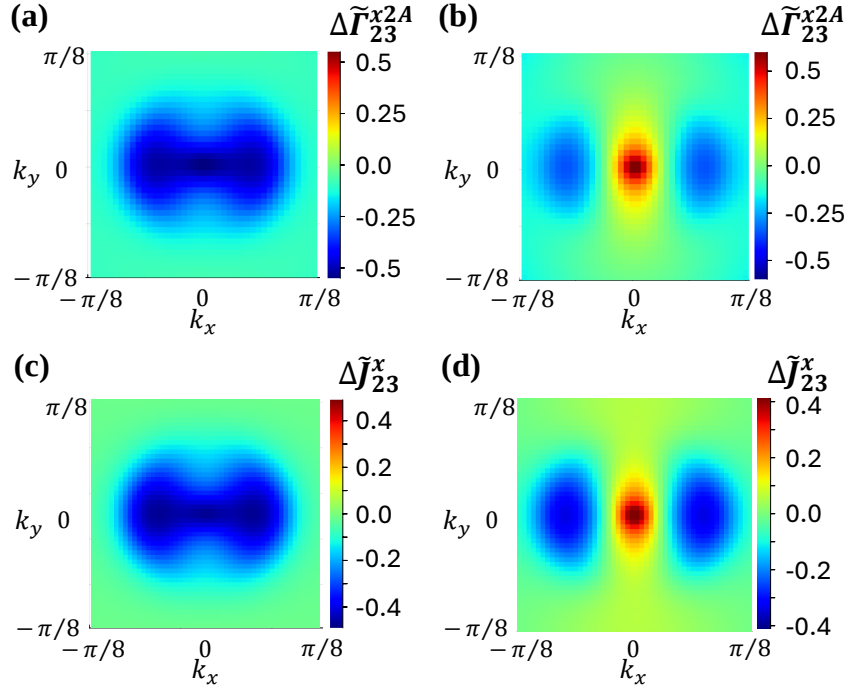


Figure 3.13: Distribution of (a) (b) $\Delta\tilde{\Gamma}_{23}^{x2A}$ and (c) (d) $\Delta\tilde{J}_{23}^x$ in the $\mathbf{k}$ space. We set the magnetic field (a) (c) $h = 0.05$ and (b) (d) $h = 0.08$ below and above the topological transition, respectively. The parameters are set as $\tilde{\mu} = 0.0$ and $U = 1.15$. From Fig. 14 of Ref. [79]. © 2025 American Physical Society

inversion at the topological transition. The eigenenergies of Bogoliubov quasiparticles $E_{2\mathbf{k}}$ and $E_{3\mathbf{k}}$ are obtained at $\mathbf{k} = \mathbf{0}$

$$(E_{2\mathbf{k}}, E_{3\mathbf{k}}) = \begin{cases} (E^{(-)}, E^{(+)}) & (\sqrt{\tilde{\mu}^2 + \Delta^2} > h) \\ (E^{(+)}, E^{(-)}) & (\sqrt{\tilde{\mu}^2 + \Delta^2} < h) \end{cases}, \quad (3.4.11)$$

where $E^{(-)}$ and $E^{(+)}$ are defined as

$$E^{(-)} = -\sqrt{\tilde{\mu}^2 + \Delta^2} + h, \quad E^{(+)} = \sqrt{\tilde{\mu}^2 + \Delta^2} - h. \quad (3.4.12)$$

The band inversion between the $E_{2\mathbf{k}}$ and $E_{3\mathbf{k}}$ bands occurs at the critical magnetic field $h_c = \sqrt{\tilde{\mu}^2 + \Delta^2}$. In other word, $E^{(-)}$ and $E^{(+)}$ are exchanged at $h = h_c$. We then analytically calculate $\Delta\tilde{\Gamma}_{23}^{x2A}(0)$. When we adopt the notation $\tilde{\Gamma}_{aa}^{\alpha 2A} = \tilde{\Gamma}_{aa}^{\alpha}(-2i\gamma)$ and $\Lambda_1^{\alpha 2A} = \Lambda_1^{\alpha}(-2i\gamma)$, $\tilde{\Gamma}_{aa}^{\alpha 2A}$ is obtained as,

$$\tilde{\Gamma}_{aa}^{\alpha 2A} = \partial_{\alpha}\xi + \partial_{\alpha}\mathbf{g} \cdot \hat{\mathbf{h}} - \frac{\Delta}{\sqrt{\tilde{\mu}^2 + \Delta^2}}\Lambda_1^{\alpha 2A} \quad (\text{for } E_a = E^{(-)}), \quad (3.4.13)$$

$$\tilde{\Gamma}_{aa}^{\alpha 2A} = \partial_{\alpha}\xi - \partial_{\alpha}\mathbf{g} \cdot \hat{\mathbf{h}} + \frac{\Delta}{\sqrt{\tilde{\mu}^2 + \Delta^2}}\Lambda_1^{\alpha 2A} \quad (\text{for } E_a = E^{(+)}). \quad (3.4.14)$$

We can verify that $\tilde{\Gamma}_{aa}^{\alpha 2A}$ at $\mathbf{k} = \mathbf{0}$ is exactly independent of $\Lambda_1^{\alpha}(-2i\gamma)$, the vertex correction as a result of the phase mode. Thus, $\Delta\tilde{\Gamma}_{23}^{x2A}(0)$ is given by

$$\Delta\tilde{\Gamma}_{23}^{\alpha 2A}(0) = \begin{cases} 2(\partial_x\mathbf{g} \cdot \hat{\mathbf{h}}) - \frac{2\Delta}{\sqrt{\tilde{\mu}^2 + \Delta^2}}\Lambda_1^{\alpha 2A} & (h_c > h) \\ -2(\partial_x\mathbf{g} \cdot \hat{\mathbf{h}}) + \frac{2\Delta}{\sqrt{\tilde{\mu}^2 + \Delta^2}}\Lambda_1^{\alpha 2A} & (h_c < h) \end{cases}. \quad (3.4.15)$$

The sign of $\Delta\tilde{\Gamma}_{23}^{\alpha 2A}(0)$ is shown to change at the topological transition.

We compare the group velocity difference $\Delta\tilde{J}_{23}^{\alpha}$ [Figs. 3.13(c) and (d)] with $\Delta\tilde{\Gamma}_{23}^{\alpha 2A}(0)$ [Figs. 3.13(a) and (b)]. A similar sign change is shown for the group velocity difference $\Delta\tilde{J}_{23}^{\alpha}$, and it is the origin of the bare photocurrent conductivity as discussed in Sec. 2.3. Note that $\Delta\tilde{J}_{23}^{\alpha}$, which we call the ‘group velocity difference’, is not exactly equal to $\partial_{k_{\alpha}}E_2 - \partial_{k_{\alpha}}E_3$ owing to the mixing of electron and hole in the Bogoliubov quasiparticle. However, it is reasonable to interpret $\Delta\tilde{\Gamma}_{23}^{\alpha 2A}(0)$ as the group velocity difference between the Bogoliubov quasiparticles, since $\tilde{J}_a^{\alpha}$ can be describe as the weighted sum of the group velocities of the electron and hole bands [57].

3.4.5 Interband pairing and quantum geometry

We have shown that the amplitude mode enhances the photocurrent conductivities in Subsect. 3.4.3. We numerically demonstrate that the interband pairing is essential for the enhancement. Furthermore, we show that the peculiar quantum geometry around the Dirac point also significantly enhances the photocurrent conductivity. The interplay between collective modes and quantum geometry results in a significant enhancement of photocurrent generation.

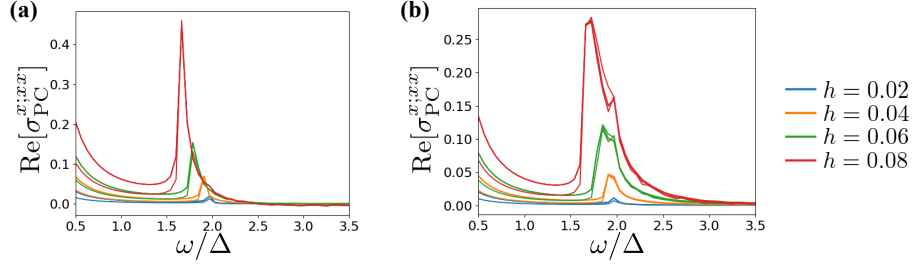


Figure 3.14: The photocurrent conductivity $\text{Re}[\sigma_{\text{PC}}^{xx}]$ in the topologically trivial states for $\tilde{\mu} = 0.2$ and $U = 1.0$. The unit of the vertical axis is $(e^3/\hbar \cdot a/\text{meV})$. Numerical results obtained by several methods are shown for each h , as in Fig. 3.11. (a) The bare photocurrent conductivity, and (b) the corrected photocurrent conductivity. From Fig. 12 of Ref. [79]. © 2025 American Physical Society

Interband pairing and enhancement by the amplitude mode

We have shown that the resonant component of the photocurrent conductivity, which exhibits a sign reversal, is enhanced by the amplitude mode. Here, we discuss the role of interband pairing for the enhancement. To verify the importance of interband pairing, we calculate the photocurrent conductivity in the case of $\tilde{\mu} = 0.2$. Although interband pairing is significant for $\tilde{\mu} = 0$, intraband pairing is dominant for $\tilde{\mu} = 0.2$. As shown in Fig. 3.14, for $\tilde{\mu} = 0.2$, the peak of the photocurrent conductivity is not enhanced but suppressed by the vertex correction in some parameter regions. Note that the sign reversal does not occur in this case because the critical magnetic field $h_c = \sqrt{\tilde{\mu}^2 + \Delta^2}$, for the topological transition, is too large to stabilize the superconducting state. In contrast to the case of $\tilde{\mu} = 0$, sharp peaks of the photocurrent conductivities are slightly suppressed by the collective modes. It indicates that the interband pairing is essential for the enhancement of the photocurrent conductivity by the amplitude mode.

Enhancement by quantum geometry

Here, we highlight the effect of quantum geometry on the photocurrent conductivity. When the Fermi level is close to the Dirac point, the JDOS $\tilde{J}_{\text{B2(ii)}}$ [Eq. (2.2.42)], which corresponds to the B2 transition around the Dirac point, gives dominant contribution to the photocurrent conductivity $\text{Re}[\sigma_{\text{PC}}^{xx}]$. The characteristic frequency dependence of $\text{Re}[\sigma_{\text{PC}}^{xx}]$ reflects the energy difference between the $E_{2\mathbf{k}}$ and $E_{3\mathbf{k}}$ bands around the Dirac point. However, the value of $\tilde{J}_{\text{B2(ii)}}$ is much smaller than $\tilde{J}_{\text{B2(i)}}$, which originates from FS1 in Fig. 3.4, and the discussion of the JDOS is not sufficient to explain the large resonant photocurrent in the low-frequency regime. We then focus on the quantum geometric properties of Bogoliubov bands, as it has been shown that quantum geometry yields colossal nonlinear

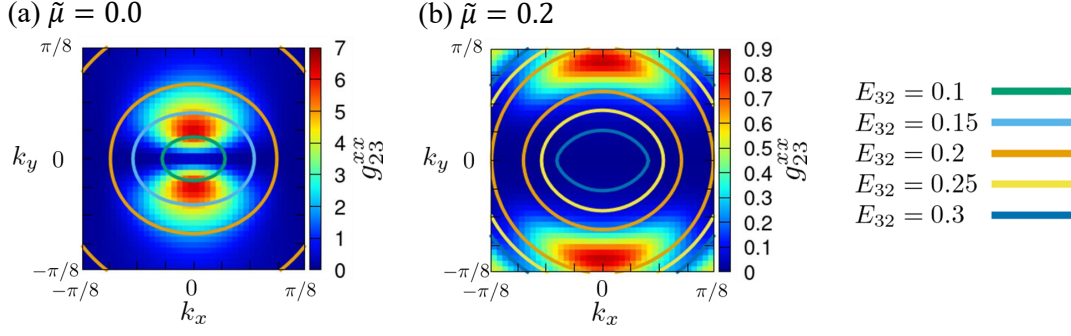


Figure 3.15: Distribution of the quantum metric g_{32}^{xx} in the $\mathbf{k}$ space. We fix $h = 0.05$, $\theta = 0^\circ$, and $\Delta = 0.09$ and set (a) $\tilde{\mu} = 0.0$ and (b) $\tilde{\mu} = 0.2$. Colored lines plot contour lines of E_{23} . Note that the mapping range of the color plot is different between the figures because the typical values of typical values of the quantum metric vastly different between (a) and (b). Reproduced from Fig. 11 of Ref. [57]. © 2024 American Physical Society

responses in some normal states [6]. Indeed, we show that quantum geometry plays a key role in enhancing the superconducting nonlinear responses.

First, we focus on the bare photocurrent conductivity. The magnetic injection current, a dominant contribution of our results, contains the quantum metric $(g_{ab}^{\alpha\beta})$, where α means λ_α for short hand notation. In particular, the contribution originating from the B2 transition is obtained as

$$\sigma_{\text{Minj(B2)}}^{x;xx} = -\frac{\pi}{4\eta} \sum_{\mathbf{k}} (\tilde{J}_{22}^x - \tilde{J}_{33}^x) g_{32}^{xx} (f_2 - f_3) \delta(\omega - E_{32}). \quad (3.4.16)$$

This formula indicates that the band-resolved quantum metric g_{32}^{xx} affects the photocurrent conductivity in the low-frequency regime. Note that the quantum metric g_{32}^{xx} is based on the λ -derivative Berry connection in Eq. (2.2.13), which is different from the k -derivative Berry connection.

We show the distribution of the quantum metric g_{32}^{xx} in the $\mathbf{k}$ space in Fig. 3.15. Here, we fix the magnitude of the pair potential as $\Delta = 0.09$. The color map shows the huge quantum metric g_{32}^{xx} around $\mathbf{k} = \mathbf{0}$, as is generally expected for Dirac electrons. As shown by the contour lines of $E_{32} = E_3 - E_2$, the quantum metric enhances the photocurrent conductivity in the low-frequency region $\omega < 2\Delta = 0.18$. These results indicate that the quantum metric selectively enhances the photocurrent conductivity originating from the JDOS $\tilde{J}_{\text{B2(ii)}}$.

When the chemical potential is far away from the Dirac point, the contribution of $\tilde{J}_{\text{B2(ii)}}$ vanishes in the low-frequency region $\omega < 2\Delta$. The quantum metric for $\tilde{\mu} = 0.2$ is shown in Fig. 3.15(c). In contrast to the case of $\tilde{\mu} = 0$, the quantum metric g_{32}^{xx} enhances the contribution of $\tilde{J}_{\text{B2(i)}}$ around $\omega = 2\Delta$, which corresponds to FS1 in Fig. 3.4. However,

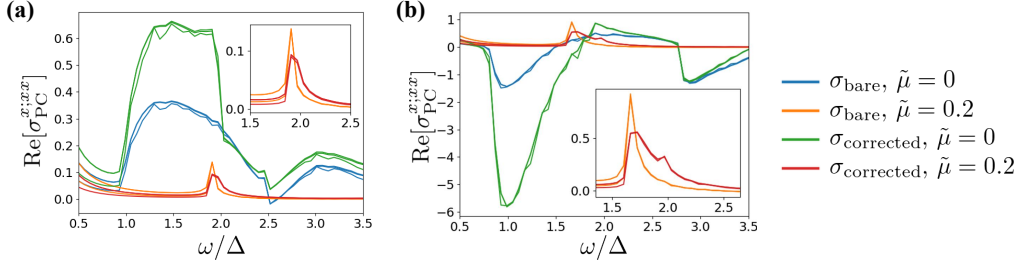


Figure 3.16: The photocurrent conductivities $\text{Re}[\sigma_{\text{PC}}^{x:xx}]$ with and without vertex corrections are compared. (a) $h = 0.04$ and (b) $h = 0.08$. The unit of the vertical axis is $(e^3/\hbar \cdot a/\text{meV})$. The parameters are set as $U = 1.0$ for $\tilde{\mu} = 0.2$ and $U = 1.15$ for $\tilde{\mu} = 0$. From Fig. 13 of Ref. [79]. © 2025 American Physical Society

the value of the photocurrent conductivity for $\tilde{\mu} = 0.2$ is smaller than that for chemical potentials near the Dirac point [compare the bare photocurrent conductivities in Figs. 3.11(a1), 3.11(b1), and 3.14(a)]

From the above results, we conclude that the quantum metric enhances the photocurrent conductivity in the low-frequency region $\omega < 2\Delta$. In the normal state, the quantum metric diverges at the Dirac point due to the energy degeneracy. Although the superconducting gap prevents the divergence of the λ -derivative quantum metric, the large quantum metric appears when the Fermi level lies near the Dirac point. It implies that the large quantum metric in the superconducting states inherits peculiar geometric properties of Dirac states.

Synergy of collective mode and quantum geometry in photocurrent generation

In the $\tilde{\mu} = 0$ case, the interband pairing is dominant and the large quantum metric appears around the Dirac point. We then focus on the interplay of collective mode and quantum geometry in the photocurrent generations. To illuminate the roles of collective modes and quantum geometry, we compare the bare and corrected photocurrent conductivities for $\tilde{\mu} = 0$ and $\tilde{\mu} = 0.2$. Figure 3.16 clearly shows that the results are qualitatively different between the cases of $\tilde{\mu} = 0$ and $\tilde{\mu} = 0.2$. For $\tilde{\mu} = 0.2$, a sharp peak of the bare conductivity is suppressed by the collective modes, while the tail in the high frequency side is enhanced. Because the interband pairing increases with increasing magnetic field, the amplitude mode is expected to enhance the photocurrent conductivity in a high frequency region $\omega/\Delta \simeq 2.0$, more significantly at high magnetic fields.

Another ingredient for the enhancement of the photocurrent conductivity is the quantum geometry. Figure 3.16 shows that the photocurrent conductivity with $\tilde{\mu} = 0$ is much larger than that with $\tilde{\mu} = 0.2$. Because the contribution of the collective modes is expected

to be of the same order as the bare photocurrent conductivity, the vertex correction is naturally enhanced by the quantum-geometric properties around the Dirac point. It indicates that the Dirac electrons are an intriguing platform for the interplay of collective modes, quantum-geometric properties, and interband pairing, leading to enhanced photocurrent generation in superconductors.

3.5 Discussion

We have demonstrated the enhancement of the linear and nonlinear optical responses due to collective modes in superconductors with Rashba spin-orbit coupling. In our calculations, the long-range Coulomb interaction was ignored. In the realistic superconductors, the massless phase mode is lifted to a high energy on the plasma frequency scale as a consequence of the Anderson-Higgs mechanism [46–49]. This change in the phase mode, due to the long-range Coulomb interaction, can affect the optical response through vertex corrections. However, the vertex correction in our model arises mainly from the amplitude (Higgs) mode, with the phase mode negligible. Therefore, the characteristic behaviors of the optical responses in our model are expected to remain unchanged even in the presence of long-range Coulomb interactions. The optical responses are enhanced by the amplitude mode, which is insusceptible to the Coulomb interaction.

3.5.1 Material platform and experiment

The sign reversal of the photocurrent conductivity is a characteristic behavior associated with the topological transition. This phenomenon can be used to verify several candidates for TSCs. The two-dimensional *s*-wave superconductor in the presence of the Rashba spin-orbit coupling and the Zeeman field, which is the setup in this study, is a platform of class D topological superconductivity [68, 71, 72]. Artificial heterostructures of an *s*-wave superconductor and a ferromagnetic insulator are proposed for an experimental platform [68–70]

In a theoretical study [121], a model of class DIII topological superconductivity also shows a sign change in the photocurrent conductivity. The mechanism of sign reversal must be different between the class D and class DIII TSCs because the magnetic injection current is absent in the latter [see Tables 2.2 and 2.1]. Although a comprehensive study is needed, sign reversal of the photocurrent conductivity may be a ubiquitous phenomenon in TSCs.

In the experiment, the sign of the injection current has been detected by direct current measurements [122, 123] and all-optical techniques [25, 86, 124]. For example, the time-resolved measurement using all-optical technique detected the electric injection current,

which grows with time in the short time pulse at the rate $\sigma_{\text{Einj}}^{\alpha;\beta\gamma}/\tau \times E^\beta E^\gamma$, in a Weyl semimetal [25]. Here, τ is the relaxation time. The injection current conductivity $\sigma_{\text{Einj}} \simeq \frac{\pi e^3}{3h^2} \times 1\text{fs}$ was observed, and τ was estimated as 8.6 fs by analyzing the Drude peak. Our numerical results show the magnitude of magnetic injection current conductivity of the order of $0.9 \times \frac{\pi e^3}{3h^2} \times \tau$ in the low-frequency region. Thus, the sign reversal behavior at the topological transition can be accessed by THz and sub-THz optical techniques.

We have shown the giant linear and nonlinear optical responses that appear when the Fermi level lies near the Dirac point. The combination of quantum geometry and collective modes plays an essential role in the enhancement. These features are expected to be realized in various quantum materials, as exemplified below. The interface of superconductors and topological insulators is an ideal platform, where superconductivity is induced in the surface Dirac/Weyl states [125, 126]. Atomically thin layered materials are another potential platform for quantum geometry and topological insulator [127, 128] and can be gate-tuned to a superconducting state [129, 130]. In few-layer WTe₂, the Berry curvature dipole was detected by the nonlinear Hall effect [131, 132]. The nonlinear Hall measurement in magnetic systems can also detect an indication for the quantum metric, and it was observed in even-layer MnBi₂Te₄ [133]. Superconductivity in MnBi₂Te₄ can be realized in the heterostructure [134]. An iron-based superconductor, FeSe, thin film is also an intriguing platform for nonlinear optical responses in superconductors because the quantum geometry of normal electrons significantly affects superconductivity [135–137].

Our results imply that the amplitude mode enhances the optical responses through interband pairing. In general, intraband pairing is expected to be favored because the interband pairing does not contribute to the thermodynamic stability of superconductivity in the weak coupling limit. One of the methods for controlling interband pairing is inducing the Zeeman field into noncentrosymmetric spin-singlet superconductors. For this purpose, two-dimensional transition metal dichalcogenides are a promising platform because the superconductivity is robust against in-plane external magnetic fields and the orbital effect is largely avoided [138–145]. Another mechanism for a significant interband pairing was proposed in noncentrosymmetric superconductors where the spin-orbit coupling has topological defects in the momentum space, such as Weyl points [76, 146].

In real superconductors, impurities diversify the mechanism of nonlinear optical responses and may complicate photocurrent conductivity. Time-resolved optical measurements, such as pump-probe spectroscopy, can monitor the ultrafast dynamics of carriers in their relaxation process [147]. Thus, we can distinguish the intrinsic contribution, such as the magnetic injection current, from those arising from impurities.

3.6 Summary

We have studied linear and nonlinear optical responses in superconductors using the self-consistent response approximation, derived from an equation of motion that conserves total particle number. Our formulas can be applied to the effective Hamiltonian describing various classes of unconventional superconductors, including anisotropic and helical superconductors. The self-consistent response approximation includes the effects of collective modes, such as amplitude and phase modes, through vertex corrections. In Secs. 3.3 and 3.4, we discussed the effects of collective modes on the linear and nonlinear conductivities. Furthermore, we extended the self-consistent response approximation to superconductors with multi-component order parameters, including spin-triplet and chiral superconductors. The Leggett mode and the Bardasis-Schrieffer (BS) mode arising from multiple Cooper channels can be appropriately accounted for in the response functions.

We applied the formulas of optical responses to a model of topological Rashba superconductors. The linear and nonlinear optical responses are strongly enhanced because of the interplay of the amplitude mode and quantum geometry when the Fermi level lies on the Dirac point. The enhancement by the amplitude mode disappears in the single-band *s*-wave superconductor, and the multiband character in the bands split by the Rashba spin-orbit coupling and the Zeeman field is essential. In particular, interband pairing is essential for the enhancement. The interplay between collective modes and quantum geometry can yield significant optical responses in topological materials, and further studies across various classes of superconductors are desired.

We have demonstrated the sign reversal of the photocurrent conductivity between the topologically trivial and nontrivial superconducting states. In our previous work, the bare photocurrent conductivity shows a sign reversal, which is attributed to magnetic injection current. Because the velocity difference between occupied and unoccupied bands induces the injection current, the magnetic injection current is sensitive to the band inversion accompanying the topological transition. It causes a sign reversal of the photocurrent conductivity. In this study, we have extended the magnetic injection current formula to include the vertex correction due to collective modes. The corrected magnetic injection current qualitatively shows the same behavior as the bare one. Consequently, the photocurrent conductivity that takes into account the vertex correction also shows the sign reversal at the topological transition. Since the self-consistent response approximation satisfies the total particle number conservation law and includes the effects of collective modes, the results in this study are more accurate than those in previous studies. Therefore, nonlinear optics would provide a bulk probe for detecting topological superconductivity in Rashba systems. The enhancement of the photocurrent conductivity by the amplitude mode is also an advantage for detecting the sign reversal behavior. The optical measurements are

expected to be a powerful bulk probe for the topological superconductivity [[57](#), [121](#), [148](#)].

Chapter 4

Valley current generation driven by surface acoustic waves in intervalley coherent states

We have mainly focused on nonlinear optical phenomena in superconductors. Superconducting states are characterized by $U(1)$ -gauge symmetry breaking, which gives rise to anomalous electromagnetic phenomena, such as the Meissner effect [55, 89]. Here, we explore exotic response phenomena arising from valley-gauge-symmetry breaking, an alternative gauge-symmetry-breaking in terms of the valley degree of freedom. Recent experiments have reported a valley-gauge-symmetry-broken phase, identified as intervalley coherent (IVC) states, in two-dimensional materials with a honeycomb structure [149–171].

We therefore focus on a pseudogauge field, an artificial gauge field arising from the lattice modulation in the honeycomb systems [172–184]. The pseudogauge field couples to the electrons like the minimal coupling, but the sign of the coupling constant is opposite between the different valley points. Thus, the pseudogauge field can be interpreted as an external valley gauge field that directly activates the valley degree of freedom. An analogy to superconductivity implies a pseudogauge-field-driven response arising from valley-gauge-symmetry breaking in the IVC states.

In this study, we investigate valley-current generation driven by surface acoustic waves (SAWs) in the IVC states [185]. SAWs modulate the two-dimensional honeycomb lattices and couple to the electrons via the pseudogauge field. We show that the IVC order gives rise to anomalous valley-current generation. The anomalous valley current exhibits a characteristic power-law dependence for low-frequency SAWs. Furthermore, we demonstrate, through numerical analysis, that the IVC order significantly enhances valley-current generation in rhombohedral graphene. These results open a pathway to explore exotic phenom-

ena arising from valley-gauge-symmetry breaking, in close analogy with gauge-symmetry breaking in superconductors.

This chapter is organized as follows. In Sec. 4.1, we explain the definition and the characteristics of the IVC states in two-dimensional honeycomb systems. In Sec. 4.2, we discuss the SAWs on the monolayer graphene and introduce a pseudogauge field. In Sec. 4.3, we discuss the similarity between the valley gauge field and the pseudogauge field and provide an overview of our expectation of anomalous valley current generation. In Sec. 4.4, we introduce a tight-binding model for rhombohedral trilayer graphene, which is one of the promising platforms for the IVC states. In Sec. 4.5, we formulate the generation of spatio-temporally uniform valley current driven by a pseudogauge field, which we refer to as the valley acoustogalvanic effect. In Sec. 4.6, we introduce the nonreciprocal pseudo-superfluid density (NRPSF) and discuss the relation between the anomalous valley current generation and the NRPSF. In Sec. 4.7, we present numerical results and verify the anomalous valley current generation arising from the intrinsic valley gauge symmetry breaking. Finally, this chapter is summarized in Sec. 4.8.

4.1 Intervalley coherent states

Various types of spin and valley ordering have recently been reported in two-dimensional materials with the honeycomb structure. Notably, the intervalley coherent (IVC) state has been focused as an exotic ordered state that spontaneously breaks valley gauge symmetry [149–171]. In this section, we will introduce the IVC states.

4.1.1 Valley degree of freedom

First, we defined the valley degree of freedom of the electron in the two-dimensional materials with the honeycomb structure. Here, we consider a monolayer graphene [Figure

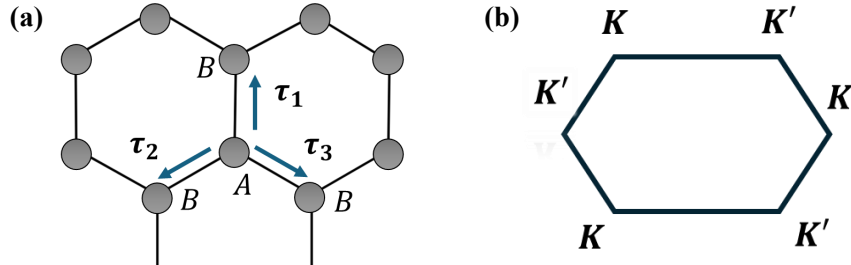


Figure 4.1: (a) The honeycomb lattice of the monolayer graphene and the hopping from the A sublattice to B sublattice. (b) The first Brillouin zone.

4.1(a)]. The hopping Hamiltonian is described as

$$\mathcal{H}^{1-\text{LG}} = \sum_{i,n} \left(c_A^\dagger(\mathbf{R}_i), c_B^\dagger(\mathbf{R}_i + \boldsymbol{\tau}_n) \right) \begin{pmatrix} 0 & -\gamma_0(\boldsymbol{\tau}_n) \\ -\gamma_0(\boldsymbol{\tau}_n) & 0 \end{pmatrix} \begin{pmatrix} c_A(\mathbf{R}_i) \\ c_B(\mathbf{R}_i + \boldsymbol{\tau}_n) \end{pmatrix}, \quad (4.1.1)$$

where $\boldsymbol{\tau}_n$ are

$$\boldsymbol{\tau}_1 = a \left(0, \frac{1}{\sqrt{3}} \right), \quad \boldsymbol{\tau}_2 = a \left(-\frac{1}{2}, -\frac{1}{2\sqrt{3}} \right), \quad \boldsymbol{\tau}_3 = a \left(\frac{1}{2}, -\frac{1}{2\sqrt{3}} \right). \quad (4.1.2)$$

We perform the Fourier transformation,

$$c_A(\mathbf{R}_i) = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} c_{A,\mathbf{k}} e^{i\mathbf{k} \cdot \mathbf{R}_i}, \quad c_B(\mathbf{R}_i + \boldsymbol{\tau}_n) = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} c_{B,\mathbf{k}} e^{i\mathbf{k} \cdot (\mathbf{R}_i + \boldsymbol{\tau}_n)}. \quad (4.1.3)$$

Then, the Hamiltonian is obtained as

$$\mathcal{H}^{1-\text{LG}} = \sum_{\mathbf{k},n} \begin{pmatrix} c_{A,\mathbf{k}}^\dagger & c_{B,\mathbf{k}}^\dagger \end{pmatrix} \begin{pmatrix} 0 & -\gamma_0(\boldsymbol{\tau}_n) e^{i\mathbf{k} \cdot \boldsymbol{\tau}_n} \\ -\gamma_0(\boldsymbol{\tau}_n) e^{-i\mathbf{k} \cdot \boldsymbol{\tau}_n} & 0 \end{pmatrix} \begin{pmatrix} c_{A,\mathbf{k}} \\ c_{B,\mathbf{k}} \end{pmatrix}. \quad (4.1.4)$$

We then focus on the $\mathbf{K} = \frac{4\pi}{3a}$ and $\mathbf{K}' = -\frac{4\pi}{3a}$, which are corners of the first Brillouin zone.

The hopping Hamiltonian around the valley points around $\mathbf{K}$ and $\mathbf{K}'$ is described as

$$\mathcal{H}_\xi^{1-\text{LG}} = \sum_{\mathbf{k},n} \begin{pmatrix} c_{A,\xi,\mathbf{k}}^\dagger & c_{B,\xi,\mathbf{k}}^\dagger \end{pmatrix} \begin{pmatrix} 0 & -\gamma_0(\boldsymbol{\tau}_n) e^{i(\xi\mathbf{K}+\mathbf{k}) \cdot \boldsymbol{\tau}_n} \\ -\gamma_0(\boldsymbol{\tau}_n) e^{-i(\xi\mathbf{K}+\mathbf{k}) \cdot \boldsymbol{\tau}_n} & 0 \end{pmatrix} \begin{pmatrix} c_{A,\xi,\mathbf{k}} \\ c_{B,\xi,\mathbf{k}} \end{pmatrix}, \quad (4.1.5)$$

where $\xi = \pm$ corresponds to valley K and K' . When we assume that the hopping parameter γ_0 does not depend on the direction $\boldsymbol{\tau}_n$ as $\gamma_0(\boldsymbol{\tau}_n) = \gamma_0$, the off-diagonal matrix element of the Hamiltonian is described as

$$-\sum_n \gamma_0(\boldsymbol{\tau}_n) e^{i(\xi\mathbf{K}+\mathbf{k}) \cdot \boldsymbol{\tau}_n} \simeq \hbar v_0 (\xi k_x - i k_y), \quad (4.1.6)$$

where the velocity is given as $\hbar v_0 = \frac{\sqrt{3}}{2} \gamma_0 a$. Therefore, the Hamiltonian of the monolayer graphene around K and K' is effectively described as a Dirac system. Because the energy bands have a Dirac cone around K and K' , these points are called valley points. The charge neutral point of the graphene lies on the Dirac point. Thus, the low-energy physics is accurately described by the effective Dirac Hamiltonian.

4.1.2 Intervalley coherence and valley gauge symmetry breaking

We have introduced the valley degree of freedom ξ of the electron $c_{\alpha,\xi,\mathbf{k}}$ based on the effective Hamiltonian. In general, the index α represents the sublattice and spin degree of freedom. The intervalley coherence is then defined as $\langle c_{\alpha,+,\mathbf{k}}^\dagger c_{\beta,-,\mathbf{k}} \rangle \neq 0$. The finite

intervalley coherence breaks the valley gauge symmetry. When considering the valley gauge transformation $c_{\alpha,\xi,\mathbf{k}} \rightarrow e^{i\xi\theta} c_{\alpha,\xi,\mathbf{k}}$, the IVC order is transformed as

$$\langle c_{\alpha,+,\mathbf{k}}^\dagger c_{\beta,-,\mathbf{k}} \rangle \rightarrow e^{-2i\xi\theta} \langle c_{\alpha,+,\mathbf{k}}^\dagger c_{\beta,-,\mathbf{k}} \rangle, \quad (4.1.7)$$

which indicates the valley gauge symmetry breaking.

The intervalley coherence is the correlation of electrons between the different valley points. The minimum difference of K and K' is $|K - K'| = \frac{4\pi}{3a}$. On the other hand, the reciprocal lattice vectors of the honeycomb lattice are given by

$$\mathbf{b}_1 = \frac{2\pi}{a} \left(1, -\frac{1}{\sqrt{3}} \right), \quad \mathbf{b}_2 = \frac{2\pi}{a} \left(0, \frac{2}{\sqrt{3}} \right). \quad (4.1.8)$$

When comparing $K - K'$ and $\mathbf{b}_i$, the ratio of the norm is obtained as $|K - K'|/|\mathbf{b}_i| = \frac{1}{\sqrt{3}}$. Indeed, the IVC order can induces the $\sqrt{3} \times \sqrt{3}$ spatial modulation with respect to the original unit cell. The spatial modulation is called Kekulé distortion. The $\sqrt{3} \times \sqrt{3}$ modulation has been detected mainly through scanning tunneling microscopy (STM) and spectroscopy (STS) experiments [149–158].

Mean-field Hamiltonian for the IVC states

We construct a mean-field Hamiltonian for the IVC states from the microscopic interaction Hamiltonian. We start from the interacting Hamiltonian,

$$\mathcal{H}_{\text{int}} = \frac{1}{2} \int d\mathbf{r}_1 \int d\mathbf{r}_2 \sum V^{\alpha\beta\gamma\delta}(\mathbf{r}_1 - \mathbf{r}_2) c_\alpha^\dagger(\mathbf{r}_1) c_\beta^\dagger(\mathbf{r}_2) c_\delta(\mathbf{r}_2) c_\gamma(\mathbf{r}_1). \quad (4.1.9)$$

The interacting Hamiltonian can be rewritten as

$$\mathcal{H}_{\text{int}} = \frac{1}{2} \sum_{\mathbf{k}_1, \mathbf{k}_2, \mathbf{q}} V^{\alpha\beta\gamma\delta}(\mathbf{q}) c_{\alpha, \mathbf{k}_1 + \mathbf{q}}^\dagger c_{\beta, \mathbf{k}_2 - \mathbf{q}}^\dagger c_{\delta, \mathbf{k}_2} c_{\gamma, \mathbf{k}_1} \quad (4.1.10)$$

$$= \frac{1}{2} \sum_{\mathbf{k}_1, \mathbf{k}_2, \mathbf{q}} V^{\alpha\beta\gamma\delta}(\mathbf{q}) c_{\alpha, \mathbf{k}_1 + \mathbf{q}}^\dagger c_{\gamma, \mathbf{k}_1} c_{\beta, \mathbf{k}_2 - \mathbf{q}}^\dagger c_{\delta, \mathbf{k}_2} + \frac{1}{2} \sum_{\mathbf{k}_1, \mathbf{k}_2, \mathbf{q}} V^{\alpha\beta\delta\gamma}(\mathbf{q}) c_{\alpha, \mathbf{k}_1}^\dagger c_{\delta, \mathbf{k}_1} c_{\beta, \mathbf{k}_2 - \mathbf{q}}^\dagger c_{\gamma, \mathbf{k}_2}, \quad (4.1.11)$$

where the Fourier component of the operators and potentials are given by

$$c_\alpha(\mathbf{r}) = \frac{1}{\sqrt{V_0}} \sum_{\mathbf{k}} c_{\alpha, \mathbf{k}} e^{i\mathbf{k} \cdot \mathbf{r}}, \quad (4.1.12)$$

$$V^{\alpha\beta\gamma\delta}(\mathbf{q}) = \frac{1}{V_0} \int d\mathbf{r} V^{\alpha\beta\gamma\delta}(\mathbf{r}) e^{-i\mathbf{q} \cdot \mathbf{r}}. \quad (4.1.13)$$

The second term in Eq. (4.1.11) is ignored in the following. Here, we focus on the intervalley scattering via $V^{\alpha\beta\gamma\delta}(\mathbf{K} - \mathbf{K}')$ [$V^{\alpha\beta\gamma\delta}(-\mathbf{K} + \mathbf{K}')$] and introduce an effective Hamiltonian,

$$\mathcal{H}_{\text{int}} = \frac{1}{2} \sum_{\mathbf{p}_1, \mathbf{p}_2} V^{\alpha\beta\gamma\delta}(\mathbf{K} - \mathbf{K}') c_{\alpha, \mathbf{K} + \mathbf{p}_1}^\dagger c_{\gamma, \mathbf{K}' + \mathbf{p}_1} c_{\beta, \mathbf{K}' + \mathbf{p}_2}^\dagger c_{\delta, \mathbf{K} + \mathbf{p}_2} \quad (4.1.14)$$

$$+ \frac{1}{2} \sum_{\mathbf{p}_1, \mathbf{p}_2} V^{\alpha\beta\gamma\delta}(-\mathbf{K} + \mathbf{K}') c_{\alpha, \mathbf{K}' + \mathbf{p}_1}^\dagger c_{\gamma, \mathbf{K} + \mathbf{p}_1} c_{\beta, \mathbf{K} + \mathbf{p}_2}^\dagger c_{\delta, \mathbf{K}' + \mathbf{p}_2}. \quad (4.1.15)$$

Introducing the IVC mean-field as

$$\Delta^{\alpha\gamma} \equiv \sum_{\mathbf{q}} V^{\alpha\beta\gamma\delta}(\mathbf{K} - \mathbf{K}') \langle c_{\beta\mathbf{K}'+\mathbf{q}}^\dagger c_{\gamma\mathbf{K}+\mathbf{q}} \rangle, \quad (4.1.16)$$

we rewrite the interaction Hamiltonian in the mean-field approximation as

$$\mathcal{H}_{\text{int}} = \sum_{\mathbf{p}} \Delta^{\alpha\beta} c_{\alpha\mathbf{K}+\mathbf{p}}^\dagger c_{\beta\mathbf{K}'+\mathbf{p}} + \Delta^{\dagger\alpha\beta} c_{\alpha\mathbf{K}'+\mathbf{p}}^\dagger c_{\beta\mathbf{K}+\mathbf{p}} + \text{const.} \quad (4.1.17)$$

The mean-field Hamiltonian is obtained as

$$\mathcal{H}_{\text{MF}} = \sum_{\mathbf{p}} \begin{pmatrix} c_{\alpha\mathbf{K}+\mathbf{p}}^\dagger & c_{\alpha\mathbf{K}'+\mathbf{p}}^\dagger \end{pmatrix} \begin{pmatrix} \mathcal{H}^{\alpha\beta}(\mathbf{K} + \mathbf{p}) & \Delta^{\alpha\beta} \\ \Delta^{\dagger\alpha\beta} & \mathcal{H}^{\alpha\beta}(\mathbf{K}' + \mathbf{p}) \end{pmatrix} \begin{pmatrix} c_{\beta\mathbf{K}+\mathbf{p}} \\ c_{\beta\mathbf{K}'+\mathbf{p}} \end{pmatrix}. \quad (4.1.18)$$

The fermion operator $c_{\alpha\mathbf{K}+\mathbf{p}}$ ($c_{\alpha\mathbf{K}'+\mathbf{p}}$) corresponds to $c_{\alpha,+,\mathbf{p}}$ ($c_{\alpha,-,\mathbf{p}}$). We later formulate the response phenomena in the IVC states based on the mean-field Hamiltonian.

Transport phenomena in the IVC states

We have explained the characteristics of the IVC states and introduced the mean-field Hamiltonian. In particular, the valley gauge symmetry breaking is an intriguing aspect of the IVC states. The analogy to superconductivity, which breaks gauge symmetry, implies the appearance of characteristic transport and response phenomena emerging from valley gauge symmetry breaking. However, only a few theoretical works have proposed the transport signature of the IVC state, such as AC Hall response under a static magnetic field [170] and characteristic magnetoresistances [171].

In this study, we show that the IVC order gives rise to anomalous valley-current generation driven by surface acoustic waves (SAWs). The anomalous valley current exhibits a characteristic power-law in SAW-frequency dependence. We verify that the exotic phenomena emerge from the valley-gauge-symmetry breaking, in close analogy with gauge-symmetry breaking in superconductors.

4.2 Surface acoustic wave

SAWs are mechanical vibrations that propagate within the unbounded surface of the elastic media [186]. When the target two-dimensional materials are placed on the elastic medium, the SAWs induce spatio-temporal modulation of the target materials. Specifically in honeycomb systems, the spatial modulation of hopping integrals can be incorporated as a pseudogauge field. The pseudogauge field affects the dynamics of the electrons [172–179]. The pseudogauge field is similar to the valley gauge field, which couples to electrons with opposite charge between the K and K' points in momentum space, unlike the standard

electromagnetic field. Thus, the pseudogauge field activates the valley degree of freedom and can drive even charge-neutral quasiparticles [180–184].

4.2.1 Pseudogauge field in a monolayer graphene

The pseudogauge fields arise from the modulation of the hopping integrals. Mechanical modulation, such as SAWs, can modulate the hopping integrals and perturb the Hamiltonian as $H_\xi(\mathbf{k}) \rightarrow H_\xi(\mathbf{k} + \frac{e}{\hbar}\xi\mathbf{A}_s)$, where $\mathbf{A}_s$ is the pseudogauge field. In this subsection, we derive the pseudogauge field for the monolayer graphene.

The hopping Hamiltonian around the valley points are described as Eq. (4.1.5). Here, we consider the effects of lattice modulation on the hopping integrals. The lattice modulation is described by the displacement field $\mathbf{u}(\mathbf{R})$ as $\mathbf{R}' = \mathbf{R} + \mathbf{u}(\mathbf{R})$. The displacement field changes the hopping vector $\boldsymbol{\tau} = \mathbf{R}_\alpha - \mathbf{R}_\beta$ as

$$\boldsymbol{\tau}' = \mathbf{R}'_\alpha - \mathbf{R}'_\beta \simeq \boldsymbol{\tau} + (\boldsymbol{\tau} \cdot \nabla)\mathbf{u}(\mathbf{R}_\alpha). \quad (4.2.1)$$

The hopping parameter is modified under the displacement field as

$$\gamma_0(\boldsymbol{\tau}') \simeq \gamma_0(\boldsymbol{\tau}) - \beta_0\gamma_0(\boldsymbol{\tau})\frac{\boldsymbol{\tau}}{\tau^2} \cdot \{(\boldsymbol{\tau} \cdot \nabla)\mathbf{u}\}, \quad (4.2.2)$$

where we assume the relation between γ_0 and τ ; $d \log \gamma_0 / d \log \tau = -\beta_0$. Thus, the off-diagonal matrix element of the hopping Hamiltonian is described with the pseudogauge field $\mathbf{A}_s$,

$$-\sum_n \gamma_0(\boldsymbol{\tau}'_n) e^{i(\xi\mathbf{K}+\mathbf{k}) \cdot \boldsymbol{\tau}'_n} \simeq \hbar v_0 [\xi(k_x + \xi A_s^x) - i(k_y + \xi A_s^y)], \quad (4.2.3)$$

The explicit form of the pseudogauge field $\mathbf{A}_s$ is obtained as

$$\begin{pmatrix} A_s^x \\ A_s^y \end{pmatrix} = -\frac{3\beta_0\gamma_0}{4\hbar v_0} \begin{pmatrix} u_{xx} - u_{yy} \\ -2u_{xy} \end{pmatrix} = -\frac{\sqrt{3}\beta_0}{2a} \begin{pmatrix} u_{xx} - u_{yy} \\ -2u_{xy} \end{pmatrix}, \quad (4.2.4)$$

where we define the strain tensor $u_{\mu\nu}$,

$$u_{\mu\nu} = \frac{1}{2} \left(\frac{\partial u_\mu}{\partial x_\nu} + \frac{\partial u_\nu}{\partial x_\mu} \right). \quad (4.2.5)$$

Consequently, we can incorporate the effect of lattice modulation on the Hamiltonian by $H_\xi(\mathbf{k}) \rightarrow H_\xi(\mathbf{k} + \frac{e}{\hbar}\xi\mathbf{A}_s)$.

Rayleigh-type SAWs propagating on the surface of piezoelectric substrates in the xy plane are described by the displacement field $\mathbf{u}(\mathbf{r}, t) = \text{Re} \left[\left(u_L \hat{Q} + i u_z \hat{z} \right) e^{i(\mathbf{Q} \cdot \mathbf{r} - \Omega t)} \right]$, where u_L and u_z are the longitudinal and out-of-plane components [187]. The SAWs propagate with the photon wave vector $\mathbf{Q} = Q(\cos \phi, \sin \phi)$ and frequency Ω . The SAW-induced pseudogauge field reads $\mathbf{A}_s = -\frac{\sqrt{3}\beta_0}{2a} u_L Q(-\cos 2\phi, \sin 2\phi) \text{Im} [e^{i(\mathbf{Q} \cdot \mathbf{r} - \Omega t)}]$.

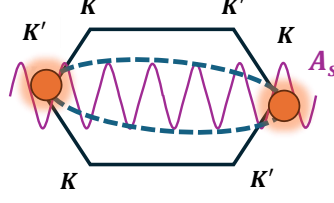


Figure 4.2: Schematic of the IVC state under the SAWs. The SAWs induce the pseudogauge field $\mathbf{A}_s$.

4.3 Valley gauge field and pseudogauge field

We briefly review the similarity between the valley gauge field and the pseudogauge field in monolayer graphene [172–176]. The monolayer graphene is described by the effective Dirac Hamiltonian $\mathcal{H}_\xi^{1-\text{LG}}(\mathbf{k})$ per valley (K/K') and spin. The valley gauge field $\mathbf{A}_v$ is incorporated into the Hamiltonian as $H_\xi^{1-\text{LG}}(\mathbf{k}) \rightarrow H_\xi^{1-\text{LG}}(\mathbf{k} + \frac{e}{\hbar}\xi\mathbf{A}_v)$. The valley gauge field, which is a counterpart of the electromagnetic field, is coupled to electrons with opposite signs between the different valleys. On the other hand, the pseudogauge field emerges from the hopping integral modulations and couples to the electron as $H_\xi^{1-\text{LG}}(\mathbf{k}) \rightarrow H_\xi^{1-\text{LG}}(\mathbf{k} + \frac{e}{\hbar}\xi\mathbf{A}_s)$. Interestingly, the two fields are formally equivalent, and the pseudogauge field $\mathbf{A}_s$ can be interpreted as an external valley gauge field.

In this thesis, we explore a pseudogauge-field driven response phenomenon arising from spontaneous valley-gauge-symmetry breaking in the IVC state [Fig. 4.2(a)]. The IVC order is a counterpart of superconductivity because the valley ($U(1)$ -) gauge symmetry is broken by the former (latter). The relation between the electromagnetic field and the $U(1)$ -gauge symmetry is essential for anomalous electromagnetic responses in superconductors, such as the Meissner effect [89]. We focus on the fact that a similar relation holds between the pseudogauge field and the valley gauge symmetry in the IVC state.

We demonstrate that the IVC order gives rise to anomalous contributions to nonlinear valley currents driven by the pseudogauge field. Furthermore, we analytically verify that the anomalous contributions arise from a novel type of macroscopic coherence, analogous to superconductivity. In the superconducting state, the superfluidity and macroscopic coherence is characterized by the superfluid density $\rho^{\alpha\beta} = \lim_{\boldsymbol{\lambda} \rightarrow \mathbf{0}} \partial_{\lambda_\alpha} \partial_{\lambda_\beta} F[\mathbf{A} = \boldsymbol{\lambda}]$, where $F_{\boldsymbol{\lambda}}$ is the free energy and $\boldsymbol{\lambda}$ is the variational parameters corresponding to the electromagnetic vector potential $\mathbf{A}$. The superfluid density determines the electromagnetic properties of superconductors. We later show that the anomalous valley current generation in the IVC state is associated with the pseudo-superfluid density, which is the derivative of the free energy with respect to the pseudogauge field. An analogy with superconductivity suggests that the valley current generation can characterize the novel macroscopic coherence of the

IVC state.

4.4 Model: rhombohedral trilayer graphene

We perform numerical calculations of SAW-driven valley-current generation in the tight-binding model of rhombohedral trilayer graphene (R3G) to demonstrate the impact of IVC ordering. R3G is one of the promising platforms for the IVC states [153, 164, 165].

4.4.1 Six-band continuum model

R3G in the normal state is accurately described using a six-band continuum model per valley (K/K') and spin [189, 190]:

$$H_{\xi}(\mathbf{k}) = \begin{pmatrix} \Delta_1 + \Delta_2 + \delta & \frac{1}{2}\gamma_2 & v_0\pi^\dagger & v_4\pi^\dagger & v_3\pi & 0 \\ \frac{1}{2}\gamma_2 & -\Delta_1 + \Delta_2 + \delta & 0 & v_3\pi^\dagger & v_4\pi & v_0\pi \\ v_0\pi & 0 & \Delta_1 + \Delta_2 & \gamma_1 & v_4\pi^\dagger & 0 \\ v_4\pi & v_3\pi & \gamma_1 & -2\Delta_2 & v_0\pi^\dagger & v_4\pi^\dagger \\ v_3\pi^\dagger & v_4\pi^\dagger & v_4\pi & v_0\pi & -2\Delta_2 & \gamma_1 \\ 0 & v_0\pi^\dagger & 0 & v_4\pi & \gamma_1 & \Delta_2 - \Delta_1 \end{pmatrix}, \quad (4.4.1)$$

where $\pi = \xi k_x + i k_y$ with $\xi = \pm$ corresponding to the valley K and K' . The Hamiltonian is written on the basis $(A_1, B_3, B_1, A_2, B_2, A_3)$, where A_i and B_i denote nonequivalent sublattice sites on layer i . The bare hopping integrals are defined as γ_i , and the band structure parameters v_i are given by $v_i = \sqrt{3}a_0\gamma_i/2$ with $i = 0, 3, 4$. The interlayer hopping processes are illustrated in Fig. 4.3(b). Intralayer hopping integral γ_0 is larger than the other interlayer hopping integrals γ_1 - γ_4 . An on-site potential δ is only present at the non-dimerized sites A_1 and B_3 . A potential difference Δ_1 between the outer layers is approximately proportional to the displacement field, while Δ_2 is a potential at the middle

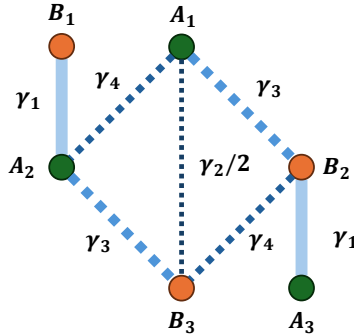


Figure 4.3: The unit cell and the interlayer hopping process in R3G. Here, A_j and B_j represent the sublattice on the layer $j = 1, 2, 3$. The hopping integrals are denoted by γ_i .

γ_0	γ_1	γ_2	γ_3	γ_4	δ	Δ_1	Δ_2	μ
3.1	0.38	-0.015	-0.29	-0.141	-0.0105	0.030	-0.0023	-0.04

Table 4.1: Tight-binding parameters and chemical potential for R3G. We adopt these values in this study. Units for all parameters are eV. The tight-binding parameters are taken from Ref. [188].

type	$\mathcal{H}_\xi^{\alpha\beta}(\mathbf{k})$	(α, β)	layer
A	$v_0\pi$	$(A_l, B_l) \quad (l = 1, 2, 3)$	intra-layer
B	const.	$(B_1, A_2), (B_2, A_3), (A_1, B_3)$	inter-layer
C1	$v_3\pi$	$(B_2, A_1), (B_3, A_2)$	inter-layer
C2	$v_4\pi$	$(A_1, A_2), (B_2, B_3)$	inter-layer

Table 4.2: Off-diagonal matrix elements of the six-band model Hamiltonian $\mathcal{H}_\xi^{\alpha\beta}(\mathbf{k})$ are classified into the types A, B, C1, and C2 based on the coupling to the pseudogauge field. The type B matrix elements are not coupled to the pseudogauge field, since they do not depend on the wave vector $\mathbf{k}$.

layer compared to the outer layers. All tight-binding parameters are chosen from Ref. [188]. We summarize the values of the tight-binding parameters and the chemical potential in Table 4.1.

4.4.2 Pseudogauge field in R3G

The SAWs generate the pseudogauge field via modulation of the hopping integrals. In monolayer graphene, where only the intralayer hopping processes occur, the pseudogauge field coincides with the valley gauge field. On the other hand, in the R3G, interlayer hopping processes are significant for electronic properties such as energy bands [189, 190], and it is necessary to consider how the pseudogauge field couples to electrons in these processes.

We derive the pseudogauge field and its coupling to the Hamiltonian from the perturbations of the tight-binding model for the R3G under the lattice modulation $\mathbf{u}(\mathbf{r}, t)$. The pseudogauge field couples to electrons like the minimal coupling, but the coupling constants depend on the hopping processes. To summarize the couplings of the pseudogauge field, we categorize the matrix elements into four types: A, B, C1, and C2, corresponding to the different hopping processes [Table 4.2]. Type A originates from intralayer hopping, while the other types correspond to interlayer hopping. The coupling to the type A elements of the Hamiltonian is the same as that to the monolayer graphene, $H_\xi^{\alpha\beta}(\mathbf{k}) \rightarrow H_\xi^{\alpha\beta}(\mathbf{k} + \frac{e}{\hbar}\xi \mathbf{A}_s)$.

Type B hopping process

Next, we consider the type B hopping process under lattice modulation. The hopping Hamiltonian is given by

$$H_B = \sum_i \left(c_\alpha^\dagger(\mathbf{R}_i), c_\beta^\dagger(\mathbf{R}_i + \boldsymbol{\tau}) \right) \begin{pmatrix} 0 & -\gamma_j(\boldsymbol{\tau}) \\ -\gamma_j(\boldsymbol{\tau}) & 0 \end{pmatrix} \begin{pmatrix} c_\alpha(\mathbf{R}_i) \\ c_\beta(\mathbf{R}_i + \boldsymbol{\tau}) \end{pmatrix}, \quad (j = 1, 2) \quad (4.4.2)$$

where the hopping direction $\boldsymbol{\tau}$ is perpendicular to the two-dimensional planes. In this study, we consider the lattice modulation propagating within the two-dimensional planes. Therefore, lattice modulation does not change the hopping parameters γ_j of type B hopping processes,

$$\gamma_j(\boldsymbol{\tau}') - \gamma_j(\boldsymbol{\tau}) \propto (\hat{z} \cdot \nabla) \mathbf{u}(x, y) = 0. \quad (4.4.3)$$

Thus, we can ignore the effect of lattice modulation on type B hopping processes.

Type C1 and C2 hopping processes

The C1 type hopping Hamiltonian is given by

$$H_{C1} = \sum_{i,n} \left(c_\alpha^\dagger(\mathbf{R}_i), c_\beta^\dagger(\mathbf{R}_i + \bar{\boldsymbol{\tau}}_n) \right) \begin{pmatrix} 0 & -\gamma_3(\bar{\boldsymbol{\tau}}_n) \\ -\gamma_3(\bar{\boldsymbol{\tau}}_n) & 0 \end{pmatrix} \begin{pmatrix} c_\alpha(\mathbf{R}_i) \\ c_\beta(\mathbf{R}_i + \bar{\boldsymbol{\tau}}_n) \end{pmatrix}. \quad (4.4.4)$$

The vector $\bar{\boldsymbol{\tau}}_n$ is defined as $\bar{\boldsymbol{\tau}}_n = \boldsymbol{\tau}_n + d\hat{z}$, where d is the distance between the layers and $\boldsymbol{\tau}_n$ is introduced by Eq. (4.1.2). The hopping parameter under lattice modulation is obtained as

$$\gamma_3(\bar{\boldsymbol{\tau}}'_n) = \gamma_3(\bar{\boldsymbol{\tau}}_n) - \beta_3 \gamma_3(\bar{\boldsymbol{\tau}}_n) \frac{\boldsymbol{\tau}_n}{\tau_n^2 + d^2} \cdot [(\boldsymbol{\tau}_n \cdot \nabla) \mathbf{u}] - \beta \gamma_3(\bar{\boldsymbol{\tau}}_n) \frac{d}{\tau_n^2 + d^2} \cdot [(\boldsymbol{\tau}_n \cdot \nabla) u_z], \quad (4.4.5)$$

where we assume the relation $d \log \gamma_3 / d \log \tau = -\beta_3$. The modulation of the hopping parameter is incorporated into the Hamiltonian as the pseudogauge field,

$$- \sum_n \gamma_3(\bar{\boldsymbol{\tau}}'_n) e^{i(\xi \mathbf{K} + \mathbf{k}) \cdot \boldsymbol{\tau}_n} \simeq \hbar v_3 [\xi \{k_x + r_3 \xi (A_s^x + \tilde{A}_s^x)\} - i \{k_y + r_3 \xi (A_s^y + \tilde{A}_s^y)\}]. \quad (4.4.6)$$

The parameter r_i , introduced as the coupling factor of the pseudogauge field $\mathbf{A}_s$, is obtained as

$$r_i = \frac{\tau^2}{\tau^2 + d^2} \cdot \frac{\beta_i}{\beta_0}. \quad (4.4.7)$$

In addition to the pseudogauge field $\mathbf{A}_s$, another pseudogauge field $\tilde{\mathbf{A}}_s$ arises into type C1 and C2 matrix elements,

$$\begin{pmatrix} \tilde{A}_s^x \\ \tilde{A}_s^y \end{pmatrix} = - \frac{3\sqrt{3}\beta_0 t}{\hbar v_3} \frac{d}{a} \begin{pmatrix} -u_{yz} \\ u_{xz} \end{pmatrix} = - \frac{6\beta_0}{a} \frac{d}{a} \begin{pmatrix} -u_{yz} \\ u_{xz} \end{pmatrix}. \quad (4.4.8)$$

We demonstrate that the second term in Eq. (4.4.8) is ignorable for the valley current generations in Appendix C. When ignoring the second term in Eq. (4.4.8), the effect of the lattice modulation is incorporated into the type C1 hopping Hamiltonian by $\mathcal{H}_\xi^{\alpha\beta}(\mathbf{k}) \rightarrow \mathcal{H}_\xi^{\alpha\beta}(\mathbf{k} + r_3\xi\mathbf{A}_s)$. Conducting a similar analysis, we can show that the type C2 hopping Hamiltonian also couples to the pseudogauge field by $\mathcal{H}_\xi^{\alpha\beta}(\mathbf{k}) \rightarrow \mathcal{H}_\xi^{\alpha\beta}(\mathbf{k} + r_4\xi\mathbf{A}_s)$.

Differences between valley gauge and pseudogauge field

The factors r_3, r_4 characterize the deviation of the pseudogauge field from the valley gauge field. In this thesis, we investigate the dependence of the valley current generation on the factors r_3, r_4 to verify that anomalous contributions originating from the IVC order exist at any r_i . In the following, we assume r_3 is equal to r_4 for simplicity and introduce the parameter $r = r_3 = r_4$. The value of r is roughly estimated as

$$r_i = \frac{a^2}{a^2 + 3d^2} \sim 0.2, \quad (4.4.9)$$

where we use $a = 2.5 \text{ \AA}$ [188] and $d = 3.4 \text{ \AA}$ [191]. We calculate the valley current generation with $0 \leq r \leq 1.2$ and verify the robustness of the anomalous valley current generation in the IVC states.

4.5 Valley acoustogalvanic effect

The oscillating pseudogauge field can drive direct electric currents via nonlinear response mechanisms, known as the acoustogalvanic effect [176, 178]. In this thesis, we focus on the generation of spatio-temporally uniform valley currents driven by a pseudogauge field, which we refer to as the valley acoustogalvanic (VAG) effect. The stationary valley current can induce valley accumulation, which is advantageous for experimental detection [192, 193]. Taking account of the pseudogauge field perturbatively, the leading order of the valley current is given by the second-order nonlinear response,

$$\langle \mathcal{J}_v^\alpha \rangle = \sigma_{s, \mathbf{Q}, -\mathbf{Q}}^{\alpha; \beta\gamma}(0; \Omega, -\Omega) E_s^\beta(\mathbf{Q}, \Omega) E_s^\gamma(-\mathbf{Q}, -\Omega), \quad (4.5.1)$$

where we introduce the Fourier components of the pseudo-electric field,

$$\mathbf{E}_s(\mathbf{r}, t) = -\partial_t \mathbf{A}_s(\mathbf{r}, t). \quad (4.5.2)$$

This formula clarifies the similarity to the nonlinear electric (NLE) conductivity,

$$\langle \mathcal{J}_e^\alpha \rangle = \sigma_{e, \mathbf{Q}, -\mathbf{Q}}^{\alpha; \beta\gamma}(0; \Omega, -\Omega) E^\beta(\mathbf{Q}, \Omega) E^\gamma(-\mathbf{Q}, -\Omega). \quad (4.5.3)$$

In the following, we assume a sufficiently large wave-length of the SAW and set $\mathbf{Q} \simeq \mathbf{0}$.

4.5.1 Formula of the valley acoustogalvanic effect

We derive the formula for the VAG effect. Our formulation is primarily based on the density-matrix method [16], which we extend for the mean-field Hamiltonian in the IVC states. The valley current operator is defined as

$$\mathcal{J}_v^\alpha = \left. \frac{\partial \mathcal{H}_{\text{MF}}(\mathbf{A}_v)}{\partial A_v^\alpha} \right|_{\mathbf{A}_v=\mathbf{0}}, \quad (4.5.4)$$

where we introduce the valley gauge field $\mathbf{A}_v$ -dependence of the Hamiltonian as

$$\mathcal{H}_{\text{MF}}(\mathbf{A}_v) = \sum_{\mathbf{k}, \alpha, \beta} \begin{pmatrix} c_{\alpha\mathbf{K}+\mathbf{k}}^\dagger & c_{\alpha\mathbf{K}'+\mathbf{k}}^\dagger \end{pmatrix} \begin{pmatrix} H_+^{\alpha\beta}(\mathbf{k} + \mathbf{A}_v) & \Delta^{\alpha\beta} \\ \Delta^{\dagger\alpha\beta} & H_-^{\alpha\beta}(\mathbf{k} - \mathbf{A}_v) \end{pmatrix} \begin{pmatrix} c_{\beta\mathbf{K}+\mathbf{k}} \\ c_{\beta\mathbf{K}'+\mathbf{k}} \end{pmatrix}. \quad (4.5.5)$$

The mean-field Hamiltonian under the SAWs is written as

$$\mathcal{H}_{\text{MF}}(\mathbf{A}_s(t)) = \sum_{\mathbf{k}, \alpha, \beta} \begin{pmatrix} c_{\alpha\mathbf{K}+\mathbf{k}}^\dagger & c_{\alpha\mathbf{K}'+\mathbf{k}}^\dagger \end{pmatrix} \begin{pmatrix} H_+^{\alpha\beta}(\mathbf{k}, \mathbf{A}_s(t)) & \Delta^{\alpha\beta} \\ \Delta^{\dagger\alpha\beta} & H_-^{\alpha\beta}(\mathbf{k}, \mathbf{A}_s(t)) \end{pmatrix} \begin{pmatrix} c_{\beta\mathbf{K}+\mathbf{k}} \\ c_{\beta\mathbf{K}'+\mathbf{k}} \end{pmatrix}. \quad (4.5.6)$$

We cannot describe the coupling to the pseudogauge field in the minimal coupling form, since the coupling constants r_i depends on the type of the hopping processes. The $\mathbf{A}_s$ -dependence of the IVC order parameter $\Delta^{\alpha\beta}$ is ignored in this study. We introduce the vertex operator to describe the formula of the VAG effects,

$$\mathcal{V}_s^\alpha = \left. \frac{\partial \mathcal{H}_{\text{MF}}(\mathbf{A}_s)}{\partial A_s^\alpha} \right|_{\mathbf{A}_s=\mathbf{0}}. \quad (4.5.7)$$

The higher-order valley current and vertex operators vanish due to the linear k -dependence of the model Hamiltonian,

$$\left. \frac{\partial^{n_1+n_2} \mathcal{H}_{\text{MF}}(\mathbf{A}_v, \mathbf{A}_s)}{\partial A_v^{\alpha_1} \dots \partial A_v^{\alpha_{n_1}} \partial A_s^{\tilde{\alpha}_1} \dots \partial A_s^{\tilde{\alpha}_{n_2}}} \right|_{\mathbf{A}_v=\mathbf{0}, \mathbf{A}_s=\mathbf{0}} = 0 \quad (n_1 + n_2 \geq 2). \quad (4.5.8)$$

In our formulation, the density matrix $\hat{\rho}(t)$ describes the physical state at time t . Without SAWs, it is written as

$$\hat{\rho}^{(0)} = \frac{e^{-\beta \mathcal{H}_{\text{MF}}(\mathbf{0})}}{\text{Tr} [e^{-\beta \mathcal{H}_{\text{MF}}(\mathbf{0})}]}, \quad (4.5.9)$$

and the time evolution of $\hat{\rho}(t)$ under SAWs is described by the von Neumann equation,

$$i\partial_t \hat{\rho}(t) = [\mathcal{H}_{\text{MF}}(\mathbf{A}_s(t)), \hat{\rho}(t)]. \quad (4.5.10)$$

Some theoretical works addressed the formula of the second-order nonlinear electric conductivity [16, 17]. We can obtain the formula of the VAG effect by replacing the electric current operators in nonlinear electric conductivity with the valley current operators or vertex operators.

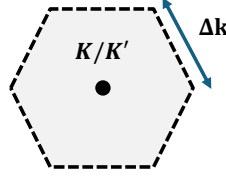


Figure 4.4: The valley charge assumed to be well-defined in the shaded domain D around the K/K' points. We parameterize the size of the domain D by Δk .

We consider the VAG conductivity $\sigma_s^{\alpha;\beta\gamma} = \sigma_{s,0,-\mathbf{0}}^{\alpha;\beta\gamma}$ in the regime $\Omega \ll \tau_{\text{rel}}^{-1} \ll \Delta E$, where τ_{rel} is the relaxation time and ΔE is the energy difference of the electron bands. We obtain the analytical formula of the VAG conductivity by generalizing the theoretical work of the NLE conductivity [16, 17]. We classify the contributions of the VAG conductivity by using the similarity to the NLE conductivity,

$$\sigma_s^{\alpha;\beta\gamma} = \sigma_{s,\text{Drude}}^{\alpha;\beta\gamma} + \sigma_{s,\text{BCP}}^{\alpha;\beta\gamma} + \sigma_{s,\text{NRSF}}^{\alpha;\beta\gamma}, \quad (4.5.11)$$

where $\sigma_{s,\text{Drude}}$, $\sigma_{s,\text{BCP}}$, and $\sigma_{s,\text{NRSF}}$ are the Drude [16, 17], Berry connection polarizability [194–196], and nonreciprocal superfluid density (NRSF) terms [16], respectively. In our model, the Berry curvature dipole term vanishes due to C_3 symmetry [18]. The variational parameters $\boldsymbol{\lambda}$ and $\tilde{\boldsymbol{\lambda}}$ are introduced as $\mathcal{H}_{\text{MF}}(\mathbf{A}_v = \boldsymbol{\lambda}, \mathbf{A}_s = \tilde{\boldsymbol{\lambda}})$ to describe the formula. The formulas are obtained as

$$\sigma_{s,\text{Drude}}^{\alpha;\beta\gamma} = \frac{1}{2(\Omega^2 + \gamma^2)} \sum_a \left[-\partial_{\epsilon_a} \partial_{\tilde{\beta}\tilde{\gamma}} f_a + \partial_{\tilde{\beta}} (\partial_{\tilde{\gamma}} f_a \cdot \partial_{\alpha} \epsilon_a) + \partial_{\tilde{\gamma}} (\partial_{\tilde{\beta}} f_a \cdot \partial_{\alpha} \epsilon_a) \right], \quad (4.5.12)$$

$$\sigma_{s,\text{BCP}}^{\alpha;\beta\gamma} = \frac{1}{4} \sum_{a,b} \left[2d_{ba}^{\omega=0} d_{aa}^{-\Omega} \epsilon_{ab} \xi_{ab}^{\alpha} \xi_{ba}^{\tilde{\beta}} \partial_{\tilde{\gamma}} f_{ba} - \xi_{ab}^{\tilde{\beta}} \xi_{ba}^{\tilde{\gamma}} d_{ab}^{\Omega} \partial_{\alpha} f_{ab} \right] + [(\tilde{\beta}, \Omega) \leftrightarrow (\tilde{\gamma}, -\Omega)], \quad (4.5.13)$$

$$\sigma_{s,\text{NRSF}}^{\alpha;\beta\gamma} = -\frac{1}{2\Omega^2} \lim_{\boldsymbol{\lambda}, \tilde{\boldsymbol{\lambda}} \rightarrow \mathbf{0}} \partial_{\alpha} \partial_{\tilde{\beta}} \partial_{\tilde{\gamma}} F[\mathbf{A}_v = \boldsymbol{\lambda}, \mathbf{A}_s = \tilde{\boldsymbol{\lambda}}], \quad (4.5.14)$$

where we use the notation $\partial_{\alpha} = \partial_{\lambda^{\alpha}}$, $\partial_{\tilde{\alpha}} = \partial_{\tilde{\lambda}^{\alpha}}$, and $d_{ab}^{\omega} = (\omega + i\gamma - \epsilon_{ab})^{-1}$. We denote the Fermi-Dirac distribution $f_a = (e^{\beta\epsilon_a} + 1)^{-1}$ and the difference $f_{ab} = f_a - f_b$. The connections ξ^{α} and $\xi^{\tilde{\alpha}}$ with geometric meaning appear in the formula of the Berry connection polarizability term, and are defined as

$$\xi_{ab}^{\alpha} = i \left\langle a_{\boldsymbol{\lambda}, \tilde{\boldsymbol{\lambda}}} \left| \frac{\partial b_{\boldsymbol{\lambda}, \tilde{\boldsymbol{\lambda}}}}{\partial \lambda^{\alpha}} \right. \right\rangle, \quad \xi_{ab}^{\tilde{\alpha}} = i \left\langle a_{\boldsymbol{\lambda}, \tilde{\boldsymbol{\lambda}}} \left| \frac{\partial b_{\boldsymbol{\lambda}, \tilde{\boldsymbol{\lambda}}}}{\partial \tilde{\lambda}^{\alpha}} \right. \right\rangle, \quad (4.5.15)$$

where $|a_{\boldsymbol{\lambda}, \tilde{\boldsymbol{\lambda}}}\rangle$ and $|b_{\boldsymbol{\lambda}, \tilde{\boldsymbol{\lambda}}}\rangle$ are eigenstates of $\mathcal{H}_{\text{MF}}(\mathbf{A}_v = \boldsymbol{\lambda}, \mathbf{A}_s = \tilde{\boldsymbol{\lambda}})$ with the eigenenergy ϵ_a and ϵ_b , respectively. In the following, we assume that the valley charge is well-defined in the k -space domain D around the valley points, and we restrict the integral to D .

4.6 Nonreciprocal pseudo-superfluid density

We discuss the NRSF term for VAG conductivity $\sigma_{s,\text{NRSF}}^{\alpha;\beta\gamma}$ focusing on its similarity to that for NLE conductivity $\sigma_{e,\text{NRSF}}^{\alpha;\beta\gamma}$ in superconductors. The superfluidity and macroscopic coherence of the superconducting state is characterized by the superfluid density $\rho^{\alpha\beta} = \lim_{\boldsymbol{\lambda} \rightarrow \mathbf{0}} \partial_{\lambda_\alpha} \partial_{\lambda_\beta} F[\mathbf{A} = \boldsymbol{\lambda}]$, where $F_{\boldsymbol{\lambda}}$ is the free energy and $\boldsymbol{\lambda}$ is the variational parameters corresponding to the electromagnetic vector potential $\mathbf{A}$. In NLE responses, the NRSF term $\sigma_{e,\text{NRSF}}$ is associated with the nonreciprocal correction to the superfluid density, which we call NRSF $f^{\alpha\beta\gamma} = \lim_{\boldsymbol{\lambda} \rightarrow \mathbf{0}} \partial_{\lambda_\alpha} \partial_{\lambda_\beta} \partial_{\lambda_\gamma} F[\mathbf{A} = \boldsymbol{\lambda}]$. The NRSF term $\sigma_{e,\text{NRSF}}$ of NLE conductivity is written by

$$\sigma_{e,\text{NRSF}}^{\alpha;\beta\gamma} = -\frac{1}{2\Omega^2} f^{\alpha\beta\gamma}. \quad (4.6.1)$$

Thus, the NRSF term originates from the unique macroscopic coherence in superconductors and is most divergent at low frequencies. In the VAG conductivity, the NRSF term is given by the nonreciprocal pseudo-superfluid density (NRPSF) $f_s^{\alpha;\beta\gamma}$,

$$\sigma_{s,\text{NRSF}}^{\alpha;\beta\gamma} = -\frac{1}{2\Omega^2} f_s^{\alpha;\beta\gamma}, \quad (4.6.2)$$

where $f_s^{\alpha;\beta\gamma}$ is defined as

$$f_s^{\alpha;\beta\gamma} = \lim_{\boldsymbol{\lambda}, \tilde{\boldsymbol{\lambda}} \rightarrow \mathbf{0}} \partial_{\lambda_\alpha} \partial_{\tilde{\lambda}_\beta} \partial_{\tilde{\lambda}_\gamma} F[\mathbf{A}_v = \boldsymbol{\lambda}, \mathbf{A}_s = \tilde{\boldsymbol{\lambda}}]. \quad (4.6.3)$$

The valley gauge field $\mathbf{A}_v$ is coupled to the Hamiltonian by $\mathcal{H}_\xi^{\alpha\beta}(\mathbf{k}) \rightarrow \mathcal{H}_\xi^{\alpha\beta}(\mathbf{k} + \frac{e}{\hbar} \xi \mathbf{A}_v)$ for all the matrix elements (α, β) , while the coupling constants of the pseudogauge field $\mathbf{A}_s$ are different between the types of matrix elements [Table 4.2] when $r \neq 1$. The similarity between Eqs. (4.6.1) and (4.6.2) demonstrates that the NRSF term in VAG conductivity represents an anomalous contribution resulting from macroscopic coherence.

The analytical formulas for the NLE and VAG conductivities are obtained using two methods: the density matrix formula and the Green's function method [16, 17]. In the Green's function method, the NRSF and NRPSF are expressed in terms of the Green's function [16]. Thus, the Green's function method can incorporate impurity scattering and many-body interactions via the self-energy. In this work, numerical calculations are performed using the density-matrix formula for computational simplicity.

Derivation of the NRSF term based on the Green's function method

Here, we formulate the VAG conductivity based on the Green's function and discuss the effects of the impurity scattering and many-body interaction on the NRSF term. The derivation is similar to that performed in Ref. [16] for the NLE conductivity. We show

the final result of the VAG conductivity,

$$\begin{aligned} \sigma_s^{\alpha;\beta\gamma}(\Omega, ; \Omega_1, \Omega_2) = & -\frac{1}{\Omega_1\Omega_2} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi i} f(\omega) \\ & \frac{1}{2} \text{Tr}[J_v^\alpha G^R(\omega + \Omega) V_s^\beta G^R(\omega + \Omega_2) V_s^\gamma (G^R(\omega) - G^A(\omega)) \\ & + J_v^\alpha G^R(\omega + \Omega_1) V_s^\beta (G^R(\omega) - G^A(\omega)) V_s^\gamma G^A(\omega - \Omega_2) \\ & + J_v^\alpha (G^R(\omega) - G^A(\omega)) V_s^\beta G^A(\omega - \Omega_1) V_s^\gamma G^A(\omega - \Omega) \\ & + [(\beta, \Omega_1) \leftrightarrow (\gamma, \Omega_2)]], \end{aligned} \quad (4.6.4)$$

where Tr represents the trace over eigenstates of the mean-field Hamiltonian. We ignore the higher-order valley current and vertex operators in the formula, since they vanish in the model for R3G. The retarded (G^R) and advanced (G^A) Green's functions are introduced as

$$G^R(\Omega) = \frac{1}{\Omega + i\eta - H_{\text{MF}}}, \quad G^A(\Omega) = (G^R(\Omega))^\dagger, \quad (4.6.5)$$

where $\eta > 0$ is an infinitesimal parameter building the causality into the Green's function. Note that we ignore impurity scattering and many-body interactions here.

Our current interest is in low-frequency behaviors. Thus, we take the $(\Omega, \Omega_1, \Omega_2)$ -expansion from the static limit with the frequency conservation $\Omega = \Omega_1 + \Omega_2$. The leading terms are obtained as

$$\sigma_s^{\alpha;\beta\gamma}(\Omega_1 + \Omega_2; \Omega_1, \Omega_2) = \frac{1}{2\Omega_1\Omega_2} A_{\alpha\beta\gamma} + \frac{1}{2\Omega_2} B_{\alpha\beta\gamma} + \frac{1}{2\Omega_1} B'_{\alpha\beta\gamma}. \quad (4.6.6)$$

The coefficient $A_{\alpha\beta\gamma}$ is given by

$$\begin{aligned} A_{\alpha\beta\gamma} = & -\int_{-\infty}^{\infty} \frac{d\omega}{2\pi i} f(\omega) \left\{ \text{Tr}[J_v^\alpha G^R(\omega) V_s^\beta G^R(\omega) V_s^\gamma (G^R(\Omega) - G^A(\Omega)) \right. \\ & + J_v^\alpha G^R(\omega) V_s^\beta (G^R(\omega) - G^A(\omega)) V_s^\gamma G^A(\omega) \\ & \left. + J_v^\alpha (G^R(\omega) - G^A(\omega)) V_s^\beta G^A(\omega) V_s^\gamma G^A(\omega)] + (\beta \leftrightarrow \gamma) \right\}. \end{aligned} \quad (4.6.7)$$

By differentiating the Green functions by the variational parameters $\boldsymbol{\lambda}$ and $\tilde{\boldsymbol{\lambda}}$ of $\mathcal{H}_{\text{MF}}(\mathbf{A}_v = \boldsymbol{\lambda}, \mathbf{A}_s = \tilde{\boldsymbol{\lambda}})$, we obtain the relation,

$$\frac{\partial G^R(\Omega)}{\partial \lambda^\alpha} = G^R(\Omega) J_v^\alpha G^R(\Omega), \quad \frac{\partial G^R(\Omega)}{\partial \tilde{\lambda}^\alpha} = G^R(\Omega) V_s^\alpha G^R(\Omega). \quad (4.6.8)$$

Using these relations, we can finally rewrite $A_{\alpha\beta\gamma}$ as

$$\begin{aligned}
 A_{\alpha\beta\gamma} &= - \int_{-\infty}^{\infty} \frac{d\Omega}{2\pi i} f(\Omega) \lim_{\boldsymbol{\lambda}, \tilde{\boldsymbol{\lambda}} \rightarrow \mathbf{0}} \partial_{\tilde{\lambda}_\beta} \partial_{\tilde{\lambda}_\gamma} \text{Tr} [J_v^\alpha (G^R(\omega) - G^A(\omega))] \\
 &= \lim_{\boldsymbol{\lambda}, \tilde{\boldsymbol{\lambda}} \rightarrow \mathbf{0}} \partial_{\tilde{\lambda}_\beta} \partial_{\tilde{\lambda}_\gamma} \text{Tr} \left[J_v^\alpha f \left(H_{\text{MF}} \left(\boldsymbol{\lambda}, \tilde{\boldsymbol{\lambda}} \right) \right) \right] \\
 &= \lim_{\boldsymbol{\lambda}, \tilde{\boldsymbol{\lambda}} \rightarrow \mathbf{0}} \partial_{\lambda_\alpha} \partial_{\tilde{\lambda}_\beta} \partial_{\tilde{\lambda}_\gamma} \left(-\frac{1}{\beta} \text{Tr} \left[\log \left(1 + e^{-\beta H_{\text{MF}}(\boldsymbol{\lambda}, \tilde{\boldsymbol{\lambda}})} \right) \right] \right) \\
 &= \lim_{\boldsymbol{\lambda}, \tilde{\boldsymbol{\lambda}} \rightarrow \mathbf{0}} \partial_{\lambda_\alpha} \partial_{\tilde{\lambda}_\beta} \partial_{\tilde{\lambda}_\gamma} F[\mathbf{A}_v = \boldsymbol{\lambda}, \mathbf{A}_s = \tilde{\boldsymbol{\lambda}}].
 \end{aligned} \tag{4.6.9}$$

Consequently, we reproduce the formula of the NRSF term [Eq. (4.6.2)].

We then discuss the effect of impurity scattering and many-body interactions on the NRSF term. In systems without strong spatial fluctuations, we can ignore the vertex correction and the momentum dependence of the self-energy. When neglecting the vertex correction, the impurity scattering and the many-body effects are incorporated as the self-energy. Thus, the Ω^{-2} frequency dependence of the NRSF term is robust if the self-energy is not divergent.

4.6.1 NRPSF and valley gauge symmetry breaking

Next, we discuss the relation between the NRPSF $f_s^{\alpha;\beta\gamma}$ and the IVC order. Before a detailed discussion, we briefly review the relation between the NRSF $f^{\alpha\beta\gamma}$ and the superconducting order. Superconducting states are described by the Bogoliubov-de Gennes (BdG) Hamiltonian,

$$H_{\text{BdG}}(\mathbf{k}, \mathbf{A}) = \begin{pmatrix} H_N(\mathbf{k} - \frac{e}{\hbar} \mathbf{A}) & \Delta_{\text{SC}} \\ \Delta_{\text{SC}}^\dagger & H_N(-\mathbf{k} - \frac{e}{\hbar} \mathbf{A})^\top \end{pmatrix}, \tag{4.6.10}$$

where the vector potential $\mathbf{A}$ is incorporated by the minimal coupling. In the normal state (Δ_{SC}), the $\mathbf{A}$ -differentiation is almost the same as the $\mathbf{k}$ -differentiation. Since the free energy is given by

$$F[\mathbf{A} = \boldsymbol{\lambda}] = -\frac{1}{\beta} \int_{\text{BZ}} \frac{d\mathbf{k}}{(2\pi)^2} \text{Tr} \left[\log \left(1 + e^{-\beta H_{\text{BdG}}(\mathbf{k}, \boldsymbol{\lambda})} \right) \right],$$

we can rewrite $f^{\alpha\beta\gamma}$ using the equivalence of $\mathbf{A}$ - and $\mathbf{k}$ - differentiations and obtain

$$f^{\alpha\beta\gamma} = \int_{\text{BZ}} \frac{d\mathbf{k}}{(2\pi)^2} \partial_{k_\alpha} (\cdots) = 0. \tag{4.6.11}$$

We used the periodicity of the Brillouin zone (BZ) in the last equation. However, the $\mathbf{A}$ - and $\mathbf{k}$ - differentiations are not equivalent in the superconducting state, and the NRSF $f^{\alpha\beta\gamma}$ can be finite [16].

A similar relation exists between the NRPSF $f_s^{\alpha;\beta\gamma}$ and the IVC order. In the normal state ($\Delta = 0$), the differentiation with the valley gauge field is equivalent to the $\mathbf{k}$ -differentiation for the mean-field Hamiltonian, Eq. (4.5.5). Thus, we can show that $f_s^{\alpha;\beta\gamma}$ almost vanishes as

$$f_s^{\alpha;\beta\gamma} = \int_D \frac{d\mathbf{k}}{(2\pi)^2} \partial_{k_\alpha}(\cdots). \quad (4.6.12)$$

We restrict the integral range to the momentum-space domain D because the valley charge is defined there [Figure 4.4]. Since the domain D does not satisfy periodicity, the NRPSF $f_s^{\alpha;\beta\gamma}$ does not exactly vanish even in the normal state. However, our numerical results later show that the magnitude of $f_s^{\alpha;\beta\gamma}$ in the normal state is tiny and become smaller as the domain D spreads. Furthermore, the valley charge is not well-defined at momenta far from the valley points. Therefore, the valley current due to the NRPSF is probably meaningless in the normal state. In the IVC state, the IVC order forbids rewriting the integrand into the total differential with $\mathbf{k}$ as in Eq. (4.6.12). Thus, a large and physically meaningful NRPSF $f_s^{\alpha;\beta\gamma}$ appears. Indeed, we numerically demonstrate that the magnitude of $f_s^{\alpha;\beta\gamma}$ in the IVC state is much larger than that in the normal state. The similarity between NRPSF and NRPSF confirms the analogy between superconductivity and IVC order.

4.7 Numerical demonstration

Based on the above analysis, we have predicted that the IVC order makes anomalous contributions to the VAG conductivity that diverge at low frequencies. This phenomenon is a counterpart of the nonlinear NLE responses in the superconducting state. Here, we numerically demonstrate the significant impact of the IVC order on the VAG effect.

The VAG conductivity satisfies a relation $\sigma_s^{x;xx} = -\sigma_s^{x;yy} = -\sigma_s^{y;xy}$, and the other components must be zero due to C_{3v} symmetry. The frequency dependence of the VAG conductivity $\sigma_s^{x;xx}$ in the IVC state ($\Delta = 30$ meV) is shown in Fig. 4.5(a). Although the typical frequency of the SAW is in the μeV range, we plot the VAG conductivity over the frequency range from 0 to meV to clarify the frequency dependence characteristic of each mechanism of the VAG effects. The Ω^{-2} -divergence is unique to the NRPSF conductivity, and thus it is the dominant VAG effect in the μeV order frequency region.

The VAG conductivity in the normal state is plotted in Fig. 4.5(b). A finite NRPSF conductivity appears and becomes dominant in the low-frequency region $\Omega \ll 1$ meV due to the Ω^{-2} -divergence. However, the magnitude of the NRPSF conductivity in the normal state is significantly smaller than that in the IVC state. Therefore, the IVC order remarkably enhances the low-frequency VAG conductivity.

The NRPSF $f_s^{\alpha;\beta\gamma}$ determines the magnitude of the NRPSF term in the VAG conductivity

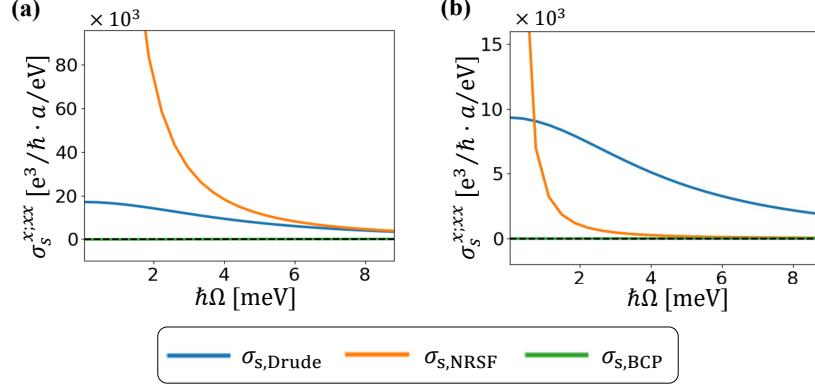


Figure 4.5: Frequency dependence of the VAG conductivity, which stems from the Drude term $\sigma_{s,\text{Drude}}$, the Berry connection polarizability term $\sigma_{s,\text{BCP}}$, and the NRSF term $\sigma_{s,\text{NRSF}}$. We set (a) $\Delta = 30 \text{ meV}$ (IVC state) and (b) $\Delta = 0 \text{ meV}$ (normal state). From Fig. 2 of Ref. [185].

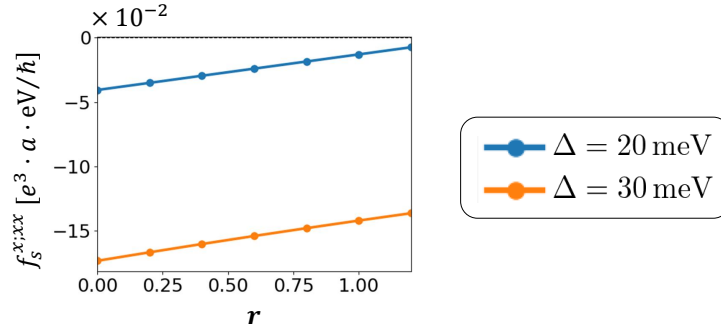


Figure 4.6: The NRPSF $f_s^{x;xx}$ as a function of r for the IVC order parameter $\Delta = 20 \text{ meV}$ and 30 meV . From Fig. 3 of Ref. [185].

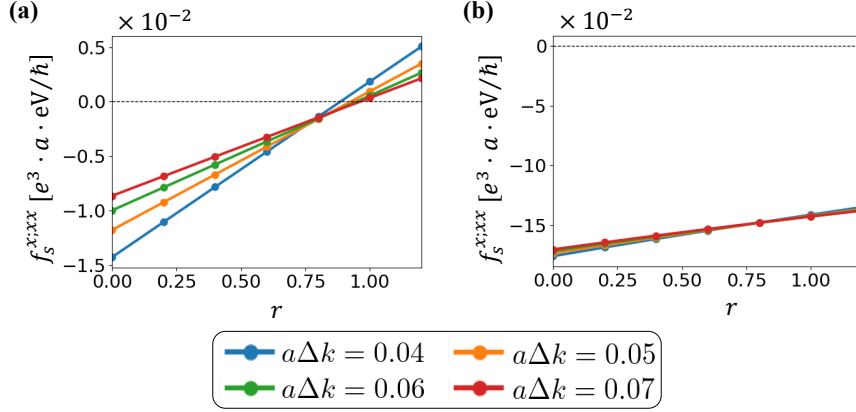


Figure 4.7: The NRPSF $f_s^{x;xx}$ for various Δk , a parameter specifying the integral domain D . We plot the r dependence in (a) the normal state ($\Delta = 0$ meV) and (b) the IVC state ($\Delta = 30$ meV). From Fig. 4 of Ref. [185].

through Eq. (4.6.2). Therefore, we here investigate how the NRPSF depends on the parameter r , which is the coupling factor for the pseudogauge field $\mathbf{A}_s$ in type C1 and C2 matrix elements [Table 4.2]. Figure 4.6 shows the r dependence of the NRPSF $f_s^{x;xx}$ with $\Delta = 20$ and 30 meV. We obtain a finite NRPSF at any r . Furthermore, the magnitude of $f_s^{x;xx}$ increases with increasing the IVC order parameter Δ . This indicates that the IVC order strongly enhances the VAG conductivity regardless of the details of the pseudogauge field.

A finite NRPSF in the normal state, Eq. (4.6.12), can be rewritten as the surface integral on the boundary of the domain D , whereas this rewriting is forbidden in the IVC state. Thus, in the normal state, the NRPSF is expected to depend heavily on the choice of the domain D , whereas in the IVC state, it is free of such ambiguity. Our numerical calculation verifies this expectation. Figure 4.7 plots the NRPSF $f_s^{x;xx}$ in the normal and IVC states with varying Δk , the parameter of the domain size D . In the normal state, the magnitude of the NRPSF decreases as the domain is enlarged. On the other hand, the NRPSF in the IVC state is almost independent of Δk . These results support our expectations. Thus, we conclude that the NRPSF is an intrinsic quantity in the IVC state, thereby verifying the exotic transport properties arising from the novel macroscopic coherence. In contrast, the NRPSF in the normal state remains as the boundary term is ambiguous because the valley charge is ill-defined far from the valley points. Thus, the NRPSF in the normal state has no physical significance. In any case, the VAG effect can indicate the IVC order via the NRPSF.

4.8 Discussion and summary

We estimate the SAW-driven valley current under a realistic experimental condition. For $\Delta \simeq 30$ meV we obtain $|f_s^{x;xx}| \simeq 0.1 e^3 \cdot a \cdot \text{eV}/\hbar$. The SAWs [197, 198] of the parameters, $u_L \simeq 2.4$ pm ($\simeq 0.8u_z$), $\beta \simeq 3$, $a \simeq 2.5\text{\AA}$, $\lambda_{\text{SAW}} = 2\pi/Q \simeq 10\mu\text{m}$ and $\Omega/2\pi \simeq 270$ MHz produce the pseudo-electric field $|\mathbf{E}_{s,0}| \simeq 20$ V/m, which gives rise to the valley current of the order of 1 A/m. When the SAW is generated by the interdigital transducers (IDTs) on the piezoelectric substrates, we can control the frequency of the SAW by varying the pitch of the IDTs [186]. The frequency dependence can be utilized to identify the Ω^{-2} -divergent contribution of the NRSF term.

Nonlocal transport measurements can detect the driven valley currents via inverse valley Hall effect [199–203]. An alternative and powerful method is Kerr rotation microscopy, one of the optical techniques [192, 193]. Circularly polarized light selectively causes the optical transition in a specific valley due to the selection rules. Therefore, valley accumulation induces Kerr rotation, and it can be used to probe the valley current. These transport and optical probes can be applied to atomic layer systems [192, 193, 199–203], platforms of the IVC order.

We comment on valley-charge conservation in the IVC state. The IVC ordering breaks the valley gauge symmetry in the mean-field theory. However, when the interaction Hamiltonian [Eq. (4.1.15)], leading to the IVC order, is valley-gauge-symmetric, valley-charge conservation holds, as proved analytically. In realistic graphene materials, intervalley scattering arises from electron-phonon coupling [204] and short-range impurity potentials [205], yet valley currents have been observed unambiguously [199–203]. Therefore, the VAG effects in the IVC states can be observed experimentally.

We have studied SAW-driven valley-current generation in the IVC state. We showed that the IVC order significantly enhances valley-current generation via valley-gauge symmetry breaking. The Ω^{-2} -divergent contribution to the VAG conductivity is attributed to the NRPSF, which is the third-order derivative of the free energy with respect to the pseudogauge and valley gauge fields. The NRPSF vanishes in the normal state when we assume the valley charge is well-defined in the whole BZ. Thus, the contribution arising from the NRPSF is unique to the valley-gauge-symmetry-broken IVC state. The SAW-driven valley-current generation highlights the analogy between the IVC order and superconductivity and may help probe IVC states. Various classes of two-dimensional materials with honeycomb lattice structure, such as rhombohedral graphene [164–168], twisted transition metal dicalcogenide [169], and twisted multilayer graphene [157–163], are candidates for the IVC state. Our results open a promising route to explore exotic response phenomena unique to valley-gauge-symmetry-broken states.

Chapter 5

Summary

In this thesis, we have investigated the nonlinear responses in exotic quantum states: the topological superconducting state and the intervalley coherent state.

In Chap. 2, we have shown the second-order nonlinear optical responses in the Rashba *s*-wave superconductors under the magnetic field, which is a promising platform for topological superconductivity. First, we verify that the longitudinal components of the nonlinear conductivity are dominantly contributed from the *k*-space region around the Dirac point by comparing the frequency dependence of the nonlinear conductivities and the joint density of states. Next, we show the sign reversal of photocurrent conductivities and the second-harmonic generation coefficient at the topological transition. By focusing on the formula for the magnetic injection current, which dominates our results, we clarify that the sign reversal is closely associated with the band inversion at the topological transition. These results imply that nonlinear optical measurements can be a powerful bulk probe of topological superconductors, as the band inversion manifests in bulk states. When demonstrating the nonlinear responses in Chap. 2, we consider quasiparticles only and ignore collective-mode excitations.

In Chap. 3, we have investigated the effects of collective modes on the optical responses in topological superconductors. Here, our methods for calculating optical responses are based on the self-consistent response approximation, which incorporates the collective modes into the linear and nonlinear conductivities via the vertex correction. We numerically showed that the amplitude mode enhances the optical responses in the Rashba superconductors when the Fermi level is close to the Dirac point. The enhancement by the amplitude mode disappears in the single-band *s*-wave superconductors. It implies that the multiband character is essential for the enhancement. Furthermore, we have demonstrated that the peculiar quantum geometry around the Dirac point also enhances the nonlinear optical conductivity. The combination of quantum geometry and collective modes plays an

important role in the significant enhancement of nonlinear optical responses. In addition, we have verified that the corrected photocurrent conductivity reproduces the sign reversal at the topological transition. These results confirm our prediction of the sign reversal in Chap. 2 and indicate that the collective mode excitation is advantageous for probing the topological superconductors.

In Chap. 4, we demonstrated anomalous valley-current generation driven by surface acoustic waves (SAWs) in the intervalley coherent (IVC) state. Intervalley coherent states are exotic states that break the valley-gauge symmetry. On the other hand, lattice modulations, such as SAWs, are approximately incorporated into the Hamiltonian as an external valley gauge field, namely a pseudogauge field, in honeycomb systems. Therefore, an analogy to superconductivity implies that the IVC order gives anomalous valley-current generation driven by SAWs. Indeed, we have verified that the valley current generation is significantly enhanced by valley-gauge-symmetry breaking and exhibits a characteristic power-law dependence on the SAW frequency. The anomalous valley current generation is closely associated with the pseudo-superfluid density, a counterpart to the superfluid density in superconductors. This is analogous to the fact that the superfluid density determines the anomalous electromagnetic properties of superconductors. Since the superfluid density characterizes the macroscopic coherence and superfluidity of superconductors, we conclude that the anomalous valley-current generation arises from the novel quantum condensation, namely, IVC order.

We have revealed the characteristics of nonlinear responses in the exotic quantum states. The remarkable nonlinear response phenomena are closely tied to the fascinating physics of quantum states, including their topological nature and intervalley coherence. Thus, the nonlinear response measurements can be a powerful tool for characterizing quantum states. Further development and exploration of nonlinear response phenomena would shed light on the hidden aspects of novel quantum materials.

Appendix A

Symmetric form

It is beneficial to rewrite the formula of the nonlinear current response function in a symmetric form, as illustrated in Fig. 3.3. Because the symmetric form does not contain the two-photon vertex explicitly, we do not have to solve the self-consistent equation for the two-photon vertex, and the computational task is significantly reduced. To derive the symmetric form, we use some useful properties of the superoperator U and Z . First, the superoperators satisfy

$$\begin{aligned} & \sum \int d\tau_1 d\tau_2 \text{Tr} \left[\overleftrightarrow{A}_{ab}(\mathbf{k}, \tau_1) \overleftrightarrow{\mathcal{G}}_{bi}(\mathbf{k}, \tau_1, \tau_2) \overleftrightarrow{U}_{ij}(\mathbf{k}, \tau_2) [B] \overleftrightarrow{\mathcal{G}}_{ja}(\mathbf{k}, \tau_2, \tau_1) \right] \\ &= \sum \int d\tau_1 d\tau_2 \text{Tr} \left[\overleftrightarrow{U}_{ab}(\mathbf{k}, \tau_1) [A] \overleftrightarrow{\mathcal{G}}_{bi}(\mathbf{k}, \tau_1, \tau_2) \overleftrightarrow{B}_{ij}(\mathbf{k}, \tau_2) \overleftrightarrow{\mathcal{G}}_{ja}(\mathbf{k}, \tau_2, \tau_1) \right], \end{aligned} \quad (\text{A.1})$$

$$\begin{aligned} & \sum \int d\tau_1 d\tau_2 \text{Tr} \left[\overleftrightarrow{A}_{ab}(\mathbf{k}, \tau_1) \overleftrightarrow{\mathcal{G}}_{bi}(\mathbf{k}, \tau_1, \tau_2) \overleftrightarrow{Z}_{ij}(\mathbf{k}, \tau_2) [B] \overleftrightarrow{\mathcal{G}}_{ja}(\mathbf{k}, \tau_2, \tau_1) \right] \\ &= \sum \int d\tau_1 d\tau_2 \text{Tr} \left[\overleftrightarrow{Z}_{ab}(\mathbf{k}, \tau_1) [A] \overleftrightarrow{\mathcal{G}}_{bi}(\mathbf{k}, \tau_1, \tau_2) \overleftrightarrow{B}_{ij}(\mathbf{k}, \tau_2) \overleftrightarrow{\mathcal{G}}_{ja}(\mathbf{k}, \tau_2, \tau_1) \right]. \end{aligned} \quad (\text{A.2})$$

The above Eqs. (A.1) and (A.2) correspond to the diagram in Figs. A.1(a) and A.1(b), respectively. Second, the superoperators satisfy the linearity,

$$\overleftrightarrow{U}_{ab}(\mathbf{k}, \tau) [X] + \overleftrightarrow{U}_{ab}(\mathbf{k}, \tau) [Y] = \overleftrightarrow{U}_{ab}(\mathbf{k}, \tau) [X + Y], \quad (\text{A.3})$$

$$\overleftrightarrow{Z}_{ab}(\mathbf{k}, \tau) [A] + \overleftrightarrow{Z}_{ab}(\mathbf{k}, \tau) [B] = \overleftrightarrow{Z}_{ab}(\mathbf{k}, \tau) [A + B]. \quad (\text{A.4})$$

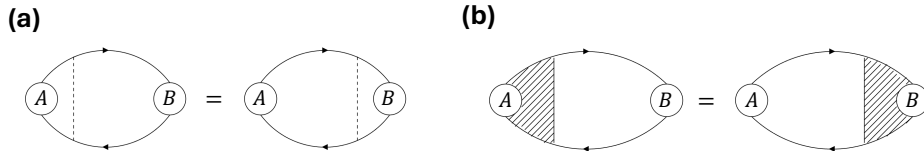


Figure A.1: The diagrammatic representation of the properties of the superoperators (a) U and (b) Z .

These properties are proved in Appendix. [A.1](#).

Using Eqs. [\(A.1\)](#) and [\(A.2\)](#), we can include a part of the two-photon vertex to correct the bare current operator. Then, we divide some terms and recollect them to construct the symmetric form using the linearity of U and Z . Finally, we get the symmetric form of the second-order nonlinear response function,

$$\begin{aligned} \tilde{\chi}^{\alpha;\beta\gamma}(\tau; \tau', \tau'') = & \frac{1}{2} \tilde{\chi}^{[\alpha\beta\gamma]}(\tau; \tau', \tau'') + \frac{1}{2} \tilde{\chi}^{[\alpha][\beta\gamma]}(\tau; \tau', \tau'') \\ & + \tilde{\chi}^{[\alpha][\beta][\gamma]}(\tau; \tau', \tau'') + \tilde{\chi}^{[\alpha\beta][\gamma]}(\tau; \tau', \tau'') + (\beta \leftrightarrow \gamma, \quad \tau' \leftrightarrow \tau''), \end{aligned} \quad (\text{A.5})$$

where we define the terms $\tilde{\chi}^{\dots}(\dots)$ as

$$\tilde{\chi}^{[\alpha\beta\gamma]}(\tau; \tau', \tau'') = \frac{1}{4} \sum \text{Tr} \left[\overleftrightarrow{J}_{ab}^{\alpha\beta\gamma}(\mathbf{k}) \overleftrightarrow{\mathcal{G}}_{ba}(\mathbf{k}, \tau^-, \tau^+) \right] \delta(\tau' - \tau') \delta(\tau' - \tau''), \quad (\text{A.6})$$

$$\begin{aligned} \tilde{\chi}^{[\alpha][\beta\gamma]}(\tau; \tau', \tau'') = & \frac{1}{4} \int d\tau_1 \sum \text{Tr} \left[\overleftrightarrow{\Gamma}_{ab}^{\alpha}(\mathbf{k}, \tau_1; \tau) \overleftrightarrow{\mathcal{G}}_{bi}(\mathbf{k}, \tau_1, \tau') \right. \\ & \times \left. \overleftrightarrow{J}_{ij}^{\beta\gamma}(\mathbf{k}) \overleftrightarrow{\mathcal{G}}_{ja}(\mathbf{k}, \tau', \tau_1) \right] \delta(\tau' - \tau''), \end{aligned} \quad (\text{A.7})$$

$$\begin{aligned} \tilde{\chi}^{[\alpha][\beta][\gamma]}(\tau; \tau', \tau'') = & \frac{1}{4} \int d\tau_1 d\tau_2 d\tau_3 \sum \text{Tr} \left[\overleftrightarrow{\Gamma}_{ab}^{\alpha}(\mathbf{k}, \tau_1; \tau) \overleftrightarrow{\mathcal{G}}_{bi}(\mathbf{k}, \tau_1, \tau_2) \overleftrightarrow{\Gamma}_{ij}^{\gamma}(\mathbf{k}, \tau_2; \tau'') \right. \\ & \times \left. \mathcal{G}_{jk}(\mathbf{k}, \tau_2, \tau_3) \overleftrightarrow{\Gamma}_{kl}^{\beta}(\mathbf{k}, \tau_3; \tau') \overleftrightarrow{\mathcal{G}}_{la}(\mathbf{k}, \tau_3, \tau_1) \right], \end{aligned} \quad (\text{A.8})$$

$$\begin{aligned} \tilde{\chi}^{[\alpha\beta][\gamma]}(\tau; \tau', \tau'') = & \frac{1}{4} \int d\tau_1 \sum \text{Tr} \left[\overleftrightarrow{J}_{ab}^{\alpha\beta}(\mathbf{k}) \overleftrightarrow{\mathcal{G}}_{bi}(\mathbf{k}, \tau, \tau_1) \right. \\ & \times \left. \overleftrightarrow{\Gamma}_{ij}^{\gamma}(\mathbf{k}, \tau_1; \tau'') \overleftrightarrow{\mathcal{G}}_{ja}(\mathbf{k}, \tau_1, \tau) \right] \delta(\tau - \tau'). \end{aligned} \quad (\text{A.9})$$

Note that the one-photon vertex function is obtained by solving the self-consistent equation, Eq. [\(3.1.51\)](#).

A.1 Properties of the superoperators

Here, we verify the properties of the superoperators [Eqs. [\(A.1\)](#)-([A.4](#))], which are used to rewrite the nonlinear response function into the symmetric form in the main text.

First, we can show the following relation of the superoperator U :

$$\begin{aligned}
 & \sum_{\mathbf{k}} \int d\tau_1 d\tau_2 \text{Tr} [\overleftrightarrow{A}_{ab}(\mathbf{k}, \tau_1) \overleftrightarrow{\mathcal{G}}_{bi}(\mathbf{k}, \tau_1, \tau_2) \overleftrightarrow{U}_{ij}(\mathbf{k}, \tau_2) [B] \overleftrightarrow{\mathcal{G}}_{ja}(\mathbf{k}, \tau_2, \tau_1)] \\
 &= -\frac{U}{V} \sum_{\mathbf{k}, \mathbf{k}'} \int d\tau_1 d\tau_2 d\bar{\tau}_1 \text{Tr} [\overleftrightarrow{A}_{ab}(\mathbf{k}, \tau_1) \overleftrightarrow{\mathcal{G}}_{bi}(\mathbf{k}, \tau_1, \tau_2) \varphi_{ij}(\mathbf{k}) \varphi_{dc}^\dagger(\mathbf{k}') \\
 & \quad \times \overleftrightarrow{P}_+ \overleftrightarrow{\mathcal{G}}_{ck}(\mathbf{k}', \tau_2, \bar{\tau}_1) \overleftrightarrow{B}_{kl}(\mathbf{k}', \bar{\tau}_1) \overleftrightarrow{\mathcal{G}}_{ld}(\mathbf{k}', \bar{\tau}_1, \tau_2) \overleftrightarrow{P}_- \overleftrightarrow{\mathcal{G}}_{ja}(\mathbf{k}, \tau_2, \tau_1)] \\
 & \quad - \frac{U}{V} \sum_{\mathbf{k}, \mathbf{k}'} \int d\tau_1 d\tau_2 d\bar{\tau}_1 \text{Tr} [\overleftrightarrow{A}_{ab}(\mathbf{k}, \tau_1) \overleftrightarrow{\mathcal{G}}_{bi}(\mathbf{k}, \tau_1, \tau_2) \varphi_{ij}^\dagger(\mathbf{k}) \varphi_{dc}(\mathbf{k}') \\
 & \quad \times \overleftrightarrow{P}_- \overleftrightarrow{\mathcal{G}}_{ck}(\mathbf{k}', \tau_2, \bar{\tau}_1) \overleftrightarrow{B}_{kl}(\mathbf{k}', \bar{\tau}_1) \overleftrightarrow{\mathcal{G}}_{ld}(\mathbf{k}', \bar{\tau}_1, \tau_2) \overleftrightarrow{P}_+ \overleftrightarrow{\mathcal{G}}_{ja}(\mathbf{k}, \tau_2, \tau_1)] \quad (\text{A.1})
 \end{aligned}$$

$$\begin{aligned}
 &= -\frac{U}{V} \sum_{\mathbf{k}, \mathbf{k}'} \int d\tau_1 d\tau_2 d\bar{\tau}_1 \text{Tr} [\varphi_{ij}(\mathbf{k}) \varphi_{dc}^\dagger(\mathbf{k}') \overleftrightarrow{P}_- \overleftrightarrow{\mathcal{G}}_{ja}(\mathbf{k}, \tau_2, \tau_1) \overleftrightarrow{A}_{ab}(\mathbf{k}, \tau_1) \overleftrightarrow{\mathcal{G}}_{bi}(\mathbf{k}, \tau_1, \tau_2) \overleftrightarrow{P}_+ \\
 & \quad \times \overleftrightarrow{\mathcal{G}}_{ck}(\mathbf{k}', \tau_2, \bar{\tau}_1) \overleftrightarrow{B}_{kl}(\mathbf{k}', \bar{\tau}_1) \overleftrightarrow{\mathcal{G}}_{ld}(\mathbf{k}', \bar{\tau}_1, \tau_2)] \\
 & \quad - \frac{U}{V} \sum_{\mathbf{k}, \mathbf{k}'} \int d\tau_1 d\tau_2 d\bar{\tau}_1 \text{Tr} [\varphi_{ij}^\dagger(\mathbf{k}) \varphi_{dc}(\mathbf{k}') \overleftrightarrow{P}_+ \overleftrightarrow{\mathcal{G}}_{ja}(\mathbf{k}, \tau_2, \tau_1) \overleftrightarrow{A}_{ab}(\mathbf{k}, \tau_1) \overleftrightarrow{\mathcal{G}}_{bi}(\mathbf{k}, \tau_1, \tau_2) \overleftrightarrow{P}_- \\
 & \quad \times \overleftrightarrow{\mathcal{G}}_{ck}(\mathbf{k}', \tau_2, \bar{\tau}_1) \overleftrightarrow{B}_{kl}(\mathbf{k}', \bar{\tau}_1) \overleftrightarrow{\mathcal{G}}_{ld}(\mathbf{k}', \bar{\tau}_1, \tau_2)] \quad (\text{A.2})
 \end{aligned}$$

$$= \sum_{\mathbf{k}'} \int d\tau_2 d\bar{\tau}_1 \text{Tr} [\overleftrightarrow{U}_{dc}(\mathbf{k}', \tau_2) [A] \overleftrightarrow{\mathcal{G}}_{ck}(\mathbf{k}', \tau_2, \bar{\tau}_1) \overleftrightarrow{B}_{kl}(\mathbf{k}', \bar{\tau}_1) \overleftrightarrow{\mathcal{G}}_{ld}(\mathbf{k}', \bar{\tau}_1, \tau_2)], \quad (\text{A.3})$$

where Tr represents the trace for the Nambu space, and $\varphi_{ab}(\mathbf{k})$ and $\varphi_{ab}^\dagger(\mathbf{k})$ act as identity operators in the Nambu space. Thus, the relation of U [Eq. (A.1)] is verified,

$$\begin{aligned}
 & \sum \int d\tau_1 d\tau_2 \text{Tr} [\overleftrightarrow{A}_{ab}(\mathbf{k}, \tau_1) \overleftrightarrow{\mathcal{G}}_{bi}(\mathbf{k}, \tau_1, \tau_2) \overleftrightarrow{U}_{ij}(\mathbf{k}, \tau_2) [B] \overleftrightarrow{\mathcal{G}}_{ja}(\mathbf{k}, \tau_2, \tau_1)] \\
 &= \sum \int d\tau_1 d\tau_2 \text{Tr} [\overleftrightarrow{U}_{ab}(\mathbf{k}, \tau_1) [A] \overleftrightarrow{\mathcal{G}}_{bi}(\mathbf{k}, \tau_1, \tau_2) \overleftrightarrow{B}_{ij}(\mathbf{k}, \tau_2) \overleftrightarrow{\mathcal{G}}_{ja}(\mathbf{k}, \tau_2, \tau_1)]. \quad (\text{A.1})
 \end{aligned}$$

Next, we derive the relation of the superoperator Z ,

$$\begin{aligned}
 & \sum \int d\tau_1 d\tau_2 \text{Tr} [\overleftrightarrow{A}_{ab}(\mathbf{k}, \tau_1) \overleftrightarrow{\mathcal{G}}_{bi}(\mathbf{k}, \tau_1, \tau_2) \overleftrightarrow{Z}_{ij}(\mathbf{k}, \tau_2) [B] \overleftrightarrow{\mathcal{G}}_{ja}(\mathbf{k}, \tau_2, \tau_1)] \\
 &= \sum \int d\tau_1 d\tau_2 \text{Tr} [\overleftrightarrow{Z}_{ab}(\mathbf{k}, \tau_1) [A] \overleftrightarrow{\mathcal{G}}_{bi}(\mathbf{k}, \tau_1, \tau_2) \overleftrightarrow{B}_{ij}(\mathbf{k}, \tau_2) \overleftrightarrow{\mathcal{G}}_{ja}(\mathbf{k}, \tau_2, \tau_1)]. \quad (\text{A.2})
 \end{aligned}$$

Here, we consider expansion of Z as a series of the superoperator U ,

$$\overleftrightarrow{Z}_{ab}(\mathbf{k}, \tau_1) [A] = \overleftrightarrow{A}_{ab}(\mathbf{k}, \tau_1) + \overleftrightarrow{Z}_{ab}^{(1)}(\mathbf{k}, \tau_1) [A] + \overleftrightarrow{Z}_{ab}^{(2)}(\mathbf{k}, \tau_1) [A] + \cdots, \quad (\text{A.4})$$

where $\overleftrightarrow{Z}_{ab}^{(i)}(\mathbf{k}, \tau_1) [A]$ is defined as

$$\overleftrightarrow{Z}_{ab}^{(0)}(\mathbf{k}, \tau_1) [A] = \overleftrightarrow{A}_{ab}(\mathbf{k}, \tau_1), \quad \overleftrightarrow{Z}_{ab}^{(n+1)}(\mathbf{k}, \tau_1) [A] = \overleftrightarrow{U}_{ab}(\mathbf{k}, \tau_1) [Z^{(n)} [A]]. \quad (\text{A.5})$$

The finite $(n+1)$ th-order term $\overleftrightarrow{Z}_{ab}^{(n+1)}(\mathbf{k}, \tau_1)[A]$ can be written as

$$\overleftrightarrow{Z}_{ab}^{(n+1)}(\mathbf{k}, \tau_1)[A] = \overleftrightarrow{U}_{ab}(\mathbf{k}, \tau_1)[U[U[\cdots U[A]\cdots]]] = \overleftrightarrow{Z}_{ab}^{(n)}(\mathbf{k}, \tau_1)[U[A]]. \quad (\text{A.6})$$

We can verify that any n -th order term of Z satisfies the relation as,

$$\begin{aligned} & \sum \int d\tau_1 d\tau_2 \text{Tr} \left[\overleftrightarrow{A}_{ab}(\mathbf{k}, \tau_1) \overleftrightarrow{\mathcal{G}}_{bi}(\mathbf{k}, \tau_1, \tau_2) \overleftrightarrow{Z}_{ij}^{(n)}(\mathbf{k}, \tau_2)[B] \overleftrightarrow{\mathcal{G}}_{ja}(\mathbf{k}, \tau_2, \tau_1) \right] \\ &= \sum \int d\tau_1 d\tau_2 \text{Tr} \left[\overleftrightarrow{Z}_{ab}^{(n)}(\mathbf{k}, \tau_1)[A] \overleftrightarrow{\mathcal{G}}_{bi}(\mathbf{k}, \tau_1, \tau_2) \overleftrightarrow{B}_{ij}(\mathbf{k}, \tau_2) \overleftrightarrow{\mathcal{G}}_{ja}(\mathbf{k}, \tau_2, \tau_1) \right], \end{aligned} \quad (\text{A.7})$$

which is proved by mathematical induction in the following way. For zeroth order, Eq. (A.7) is the same as Eq. (A.1), and thus, it is true. Now we assume that Eq. (A.7) is true for n th order. Let us check for the next $(n+1)$ th order. We can rewrite the left hand side of Eq. (A.7) for $(n+1)$ th order as

$$\begin{aligned} & \sum \int d\tau_1 d\tau_2 \text{Tr} \left[\overleftrightarrow{A}_{ab}(\mathbf{k}, \tau_1) \overleftrightarrow{\mathcal{G}}_{bi}(\mathbf{k}, \tau_1, \tau_2) \overleftrightarrow{Z}_{ij}^{(n+1)}(\mathbf{k}, \tau_2)[B] \overleftrightarrow{\mathcal{G}}_{ja}(\mathbf{k}, \tau_2, \tau_1) \right] \\ &= \sum \int d\tau_1 d\tau_2 \text{Tr} \left[\overleftrightarrow{A}_{ab}(\mathbf{k}, \tau_1) \overleftrightarrow{\mathcal{G}}_{bi}(\mathbf{k}, \tau_1, \tau_2) \overleftrightarrow{U}_{ij}(\mathbf{k}, \tau_2)[Z^{(n)}[B]] \overleftrightarrow{\mathcal{G}}_{ja}(\mathbf{k}, \tau_2, \tau_1) \right] \end{aligned} \quad (\text{A.8})$$

$$= \sum \int d\tau_1 d\tau_2 \text{Tr} \left[\overleftrightarrow{U}_{ab}(\mathbf{k}, \tau_1)[A] \overleftrightarrow{\mathcal{G}}_{bi}(\mathbf{k}, \tau_1, \tau_2) \overleftrightarrow{Z}_{ij}^{(n)}(\mathbf{k}, \tau_2)[B] \overleftrightarrow{\mathcal{G}}_{ja}(\mathbf{k}, \tau_2, \tau_1) \right] \quad (\text{A.9})$$

$$= \sum \int d\tau_1 d\tau_2 \text{Tr} \left[\overleftrightarrow{Z}_{ab}^{(n)}(\mathbf{k}, \tau_1)[U[A]] \overleftrightarrow{\mathcal{G}}_{bi}(\mathbf{k}, \tau_1, \tau_2) \overleftrightarrow{B}_{ij}(\mathbf{k}, \tau_2) \overleftrightarrow{\mathcal{G}}_{ja}(\mathbf{k}, \tau_2, \tau_1) \right] \quad (\text{A.10})$$

$$= \sum \int d\tau_1 d\tau_2 \text{Tr} \left[\overleftrightarrow{Z}_{ab}^{(n+1)}(\mathbf{k}, \tau_1)[A] \overleftrightarrow{\mathcal{G}}_{bi}(\mathbf{k}, \tau_1, \tau_2) \overleftrightarrow{B}_{ij}(\mathbf{k}, \tau_2) \overleftrightarrow{\mathcal{G}}_{ja}(\mathbf{k}, \tau_2, \tau_1) \right]. \quad (\text{A.11})$$

By mathematical induction, Eq. (A.7) is proved for arbitrary order. Therefore, the relation of the superoperator Z [Eq. (A.2)] is true.

Next, we verify the linearity of Z ,

$$\overleftrightarrow{Z}_{ab}(\mathbf{k}, \tau)[A] + \overleftrightarrow{Z}_{ab}(\mathbf{k}, \tau)[B] = \overleftrightarrow{Z}_{ab}(\mathbf{k}, \tau)[A + B]. \quad (\text{A.4})$$

We consider the self-consistent equation for some operators $\overleftrightarrow{A}_{ab}(\mathbf{k}, \tau)$ and $\overleftrightarrow{B}_{ab}(\mathbf{k}, \tau)$, by which $\overleftrightarrow{Z}_{ab}(\mathbf{k}, \tau)[A]$ and $\overleftrightarrow{Z}_{ab}(\mathbf{k}, \tau)[B]$ are determined by

$$\overleftrightarrow{Z}_{ab}(\mathbf{k}, \tau)[A] = \overleftrightarrow{A}_{ab}(\mathbf{k}, \tau) + \overleftrightarrow{U}_{ab}(\mathbf{k}, \tau)[Z[A]], \quad (\text{A.12})$$

$$\overleftrightarrow{Z}_{ab}(\mathbf{k}, \tau)[B] = \overleftrightarrow{B}_{ab}(\mathbf{k}, \tau) + \overleftrightarrow{U}_{ab}(\mathbf{k}, \tau)[Z[B]]. \quad (\text{A.13})$$

On the other hand, $\overleftrightarrow{Z}_{ab}(\mathbf{k}, \tau)[A + B]$ is given by

$$\overleftrightarrow{Z}_{ab}(\mathbf{k}, \tau)[A + B] = \overleftrightarrow{A}_{ab}(\mathbf{k}, \tau) + \overleftrightarrow{B}_{ab}(\mathbf{k}, \tau) + \overleftrightarrow{U}_{ab}(\mathbf{k}, \tau)[Z[A + B]]. \quad (\text{A.14})$$

Using the linearity of the superoperator U ,

$$\overleftrightarrow{U}_{ab}(\mathbf{k}, \tau)[X] + \overleftrightarrow{U}_{ab}(\mathbf{k}, \tau)[Y] = \overleftrightarrow{U}_{ab}(\mathbf{k}, \tau)[X + Y], \quad (\text{A.15})$$

we can rewrite the sum of $Z[A]$ and $Z[B]$ as

$$\overleftrightarrow{Z}_{ab}(\mathbf{k}, \tau)[A] + \overleftrightarrow{Z}_{ab}(\mathbf{k}, \tau)[B] = \overleftrightarrow{A}_{ab}(\mathbf{k}, \tau) + \overleftrightarrow{B}_{ab}(\mathbf{k}, \tau) + \overleftrightarrow{U}_{ab}(\mathbf{k}, \tau)[Z[A] + Z[B]]. \quad (\text{A.16})$$

Comparing Eqs. (A.14) and (A.16), the linearity of Z [Eq. (A.4)] is proved.

Appendix B

Dependence of VAG effects on propagating direction of SAWs

We consider Rayleigh-type SAWs propagating on the surface of piezoelectric substrates in the xy plane, which are described by the displacement field,

$$\mathbf{u}(\mathbf{r}, t) = \text{Re} \left[\left(u_L \hat{Q} + i u_z \hat{z} \right) e^{i(\mathbf{Q} \cdot \mathbf{r} - \Omega t)} \right]. \quad (\text{B.1})$$

Here, u_L and u_z are the longitudinal and out-of-plane components. The SAWs propagate with the phonon wave vector $\mathbf{Q} = Q(\cos \phi, \sin \phi)$ and frequency Ω . We discuss the anisotropy of the driven valley currents with respect to the propagation angle ϕ of SAWs.

The effects of lattice modulation are incorporated as pseudogauge fields. The pseudogauge field is obtained as

$$\begin{pmatrix} A_s^x \\ A_s^y \end{pmatrix} = -\frac{\sqrt{3}\beta_0}{2a} \begin{pmatrix} u_{xx} - u_{yy} \\ -2u_{xy} \end{pmatrix}. \quad (\text{B.2})$$

In addition, another pseudogauge field $\tilde{\mathbf{A}}_s$ arises in C1 and C2 type hopping Hamiltonian: $\mathcal{H}_\xi^{\alpha\beta}(\mathbf{k}) \rightarrow \mathcal{H}_\xi^{\alpha\beta}(\mathbf{k} + r_i \xi \tilde{\mathbf{A}}_s)$, and it is obtained as

$$\begin{pmatrix} \tilde{A}_s^x \\ \tilde{A}_s^y \end{pmatrix} = -\frac{6\beta_0}{a} \frac{d}{a} \begin{pmatrix} -u_{yz} \\ u_{xz} \end{pmatrix}. \quad (\text{B.3})$$

Although we ignore $\tilde{\mathbf{A}}_s$ in the main text, we here consider the contribution to the VAG effect by $\tilde{\mathbf{A}}_s$.

The pseudo-electric field $\mathbf{E}_s = -\partial_t \mathbf{A}_s$ is obtained in the main text as

$$\begin{pmatrix} E_s^x \\ E_s^y \end{pmatrix} = \frac{\sqrt{3}\beta_0}{2a} u_L Q \Omega \cdot \text{Re} \left[e^{i(\mathbf{Q} \cdot \mathbf{r} - \Omega t)} \right] \begin{pmatrix} -\cos 2\phi \\ \sin 2\phi \end{pmatrix}, \quad (\text{B.4})$$

while the additional pseudo-electric field $\tilde{\mathbf{E}}_s = -\partial_t \tilde{\mathbf{A}}_s$ arises as

$$\begin{pmatrix} \tilde{E}_s^x \\ \tilde{E}_s^y \end{pmatrix} = \frac{3\beta_0}{a} \frac{d}{a} u_z Q \Omega \cdot \text{Im} \left[e^{i(\mathbf{Q} \cdot \mathbf{r} - \Omega t)} \right] \begin{pmatrix} -\sin \phi \\ \cos \phi \end{pmatrix}. \quad (\text{B.5})$$

The total pseudo-electric field is represented as

$$[\mathbf{E}_s(\mathbf{Q}, \Omega) + i\tilde{\mathbf{E}}_s(\mathbf{Q}, \Omega)]e^{i(\mathbf{Q} \cdot \mathbf{r} - \Omega t)} + \text{c.c.}, \quad (\text{B.6})$$

with $\mathbf{E}_s(\mathbf{Q}, \Omega)$ and $\tilde{\mathbf{E}}_s(\mathbf{Q}, \Omega)$ defined as follows.

$$\mathbf{E}_s(\mathbf{Q}, \Omega) = \frac{\sqrt{3}\beta_0}{4a} u_L Q \Omega \begin{pmatrix} -\cos 2\phi \\ \sin 2\phi \end{pmatrix}, \quad \tilde{\mathbf{E}}_s(\mathbf{Q}, \Omega) = -\frac{3\beta_0}{2a} \frac{d}{a} u_z Q \Omega \begin{pmatrix} -\sin \phi \\ \cos \phi \end{pmatrix}. \quad (\text{B.7})$$

We then introduce a unified notation $E^{\bar{\alpha}}$ ($\bar{\alpha} = x, y, \tilde{x}, \tilde{y}$) such that

$$\begin{pmatrix} E^x \\ E^y \\ E^{\tilde{x}} \\ E^{\tilde{y}} \end{pmatrix} = \begin{pmatrix} E_s^x \\ E_s^y \\ \tilde{E}_s^x \\ \tilde{E}_s^y \end{pmatrix}. \quad (\text{B.8})$$

The valley current generated by the VAG effect is written as

$$J_v^\alpha(0) = \sigma^{\alpha; \bar{\beta}\bar{\gamma}}(0; \Omega, -\Omega) E^{\bar{\beta}}(\Omega) E^{\bar{\gamma}}(\Omega) + \sigma^{\alpha; \bar{\beta}\bar{\gamma}}(0; -\Omega, \Omega) E^{\bar{\beta}}(-\Omega) E^{\bar{\gamma}}(\Omega) \quad (\text{B.9})$$

$$= 2\text{Re}[\sigma^{\alpha; \bar{\beta}\bar{\gamma}}(0; \Omega, -\Omega)] \text{Re}[E^{\bar{\beta}}(\Omega) E^{\bar{\gamma}}(-\Omega)] - 2\text{Im}[\sigma^{\alpha; \bar{\beta}\bar{\gamma}}(0; \Omega, -\Omega)] \text{Im}[E^{\bar{\beta}}(\Omega) E^{\bar{\gamma}}(-\Omega)] \quad (\text{B.10})$$

$$= \text{Re} \left[\sigma^{\alpha; \bar{\beta}\bar{\gamma}}(0; \Omega, -\Omega) + \sigma^{\alpha; \bar{\gamma}\bar{\beta}}(0; \Omega, -\Omega) \right] \text{Re}[E^{\bar{\beta}}(\Omega) E^{\bar{\gamma}}(-\Omega)] \quad (\text{B.11})$$

$$- \text{Im} \left[\sigma^{\alpha; \bar{\beta}\bar{\gamma}}(0; \Omega, -\Omega) - \sigma^{\alpha; \bar{\gamma}\bar{\beta}}(0; \Omega, -\Omega) \right] \text{Im}[E^{\bar{\beta}}(\Omega) E^{\bar{\gamma}}(-\Omega)], \quad (\text{B.12})$$

where we used the relation;

$$\sigma^{\alpha; \beta\gamma}(0; \Omega, -\Omega) = \sigma^{\alpha; \gamma\beta}(0; -\Omega, \Omega), \quad (\text{B.13})$$

since the antisymmetric components do not contribute to the VAG effect.

In Chap. 4, the point group symmetry of the model for R3G is C_{3v} . The symmetry restricts the VAG conductivities,

$$\sigma^{x; x_1 x_2} = -\sigma^{x; y_1 y_2} = -\sigma^{y; x_1 y_2} = -\sigma^{y; y_1 x_2}, \quad (\text{B.14})$$

$$\sigma^{y; y_1 y_2} = -\sigma^{y; x_1 x_2} = -\sigma^{x; x_1 y_2} = -\sigma^{x; y_1 x_2} = 0, \quad (\text{B.15})$$

where the notation (x_i, y_i) is (x, y) or $(\tilde{x}, \tilde{y})$. Since the spatio-temporally uniform valley current $J_v(0)$ is a real number, we obtain the following relations

$$\sigma^{\alpha; \bar{\beta}\bar{\gamma}}(0, \Omega, -\Omega) = \left[\sigma^{\alpha; \bar{\beta}\bar{\gamma}}(0, -\Omega, \Omega) \right]^* = \left[\sigma^{\alpha; \bar{\gamma}\bar{\beta}}(0, \Omega, -\Omega) \right]^*, \quad (\text{B.16})$$

where we use Eq. (B.13). Thus, we obtain the results,

$$\text{Im} \left[\sigma^{\alpha;\bar{\beta}\bar{\gamma}}(0; \Omega, -\Omega) - \sigma^{\alpha;\bar{\gamma}\bar{\beta}}(0; \Omega, -\Omega) \right] = 0. \quad (\text{B.17})$$

Given Eqs. (B.14), (B.15), and (B.17), the valley current is given by

$$\begin{aligned} J_v^\alpha(0) = & 2\text{Re} \left[\sigma^{\alpha;\beta\gamma}(0; \Omega, -\Omega) \right] \text{Re}[E_s^\beta(\Omega)E_s^\gamma(-\Omega)] \\ & + 2\text{Re} \left[\sigma^{\alpha;\tilde{\beta}\tilde{\gamma}}(0; \Omega, -\Omega) \right] \text{Re}[\tilde{E}_s^\beta(\Omega)\tilde{E}_s^\gamma(-\Omega)] \end{aligned} \quad (\text{B.18})$$

$$= J_{v,0}^\alpha(0) + \tilde{J}_v^\alpha(0), \quad (\text{B.19})$$

where we divide the valley current into contributions $J_{v,0}^\alpha(0)$ and $\tilde{J}_v^\alpha(0)$, respectively.

We discuss how the driven valley currents depend on the propagation angle ϕ of SAWs. The $\mathbf{E}_s$ -driven valley current $J_{v,0}^\alpha$ is obtained as

$$J_{v,0}^x(0) = 2\sigma_s^{x;xx}(0; \Omega, -\Omega)(|E_s^x(\Omega)|^2 - |E_s^y(\Omega)|^2) + (\Omega \leftrightarrow -\Omega) \quad (\text{B.20})$$

$$= 2|\mathbf{E}_s(\Omega)|^2 \sigma_s^{x;xx}(\Omega) \cos 4\phi, \quad (\text{B.21})$$

$$J_{v,0}^y(0) = 2\sigma_s^{y;xy}(0; \Omega, -\Omega)(E_s^x(\Omega)E_s^y(-\Omega) + E_s^y(\Omega)E_s^x(-\Omega)) + (\Omega \leftrightarrow -\Omega) \quad (\text{B.22})$$

$$= 2|\mathbf{E}_s(\Omega)|^2 \sigma_s^{x;xx}(\Omega) \sin 4\phi. \quad (\text{B.23})$$

We then rewrite the valley current as $\mathbf{J}_{v,0} = J_{v,0}^L \hat{\mathbf{Q}} + J_{v,0}^T \hat{\boldsymbol{\phi}}$, where we introduce $\hat{\boldsymbol{\phi}} = \partial_\phi \hat{\mathbf{Q}} = (-\sin \phi, \cos \phi)$. The longitudinal and transverse components, $J_{v,0}^L$ and $J_{v,0}^T$, are given by

$$\begin{pmatrix} J_{v,0}^L \\ J_{v,0}^T \end{pmatrix} = 2|\mathbf{E}_s(\Omega)|^2 \sigma_s^{x;xx}(0; \Omega, -\Omega) \begin{pmatrix} \cos 3\phi \\ \sin 3\phi \end{pmatrix}. \quad (\text{B.24})$$

On the other hand, the $\tilde{\mathbf{E}}_s$ -driven valley current $\tilde{J}_v^\alpha$ is obtained as

$$\tilde{J}_v^x(0) = 2\sigma_s^{x;\tilde{x}\tilde{x}}(0; \Omega, -\Omega)(|\tilde{E}_s^x(\Omega)|^2 - |\tilde{E}_s^y(\Omega)|^2) \quad (\text{B.25})$$

$$= -2|\tilde{\mathbf{E}}_s(\Omega)|^2 \sigma_s^{x;\tilde{x}\tilde{x}}(0; \Omega, -\Omega) \cos 2\phi, \quad (\text{B.26})$$

$$\tilde{J}_v^\alpha(0) = 2\sigma_s^{y;\tilde{x}\tilde{y}}(0; \Omega, -\Omega)(\tilde{E}_s^x(\Omega)\tilde{E}_s^y(-\Omega) + \tilde{E}_s^y(\Omega)\tilde{E}_s^x(-\Omega)) \quad (\text{B.27})$$

$$= 2|\tilde{\mathbf{E}}_s(\Omega)|^2 \sigma_s^{x;\tilde{x}\tilde{x}}(0; \Omega, -\Omega) \sin 2\phi. \quad (\text{B.28})$$

The longitudinal and transverse components, $\tilde{J}_v^L$ and $\tilde{J}_v^T$, are given by

$$\begin{pmatrix} \tilde{J}_v^L \\ \tilde{J}_v^T \end{pmatrix} = 2|\tilde{\mathbf{E}}_s(\Omega)|^2 \sigma_s^{x;\tilde{x}\tilde{x}}(0; \Omega, -\Omega) \begin{pmatrix} -\cos 3\phi \\ \sin 3\phi \end{pmatrix}. \quad (\text{B.29})$$

Thus, we can distinguish between the $\mathbf{E}_s$ -driven and $\tilde{\mathbf{E}}_s$ -driven valley currents by the dependence on the SAW's propagation direction.

Appendix C

Estimation of valley currents driven by the additional pseudogauge field

In Appendix B, we have shown that the VAG effects can be decomposed into the $\mathbf{A}_s$ -driven and $\tilde{\mathbf{A}}_s$ -driven contributions. We then estimate the magnitude of $\tilde{J}_v^\alpha$, the valley current driven by the additional pseudogauge field $\tilde{\mathbf{A}}_s$. The couplings of Hamiltonian to $\tilde{\mathbf{A}}_s$ are summarized in Table C.1. Here, we introduce the vertex operator $\tilde{\mathcal{V}}_s^\alpha$ to formulate the $\tilde{\mathbf{A}}_s$ -driven VAG conductivity $\sigma_s^{x;\tilde{x}\tilde{x}}$. The vertex operator $\tilde{\mathcal{V}}_s^\alpha$ is defined as

$$\tilde{\mathcal{V}}_s^\alpha = \left. \frac{\partial \mathcal{H}_{\text{MF}}(\tilde{\mathbf{A}}_s, r)}{\partial \tilde{A}_s^\alpha} \right|_{\tilde{\mathbf{A}}_s=0}. \quad (\text{C.1})$$

We explicitly write the parameter r dependence with assumption $r = r_3 = r_4$. The $\tilde{\mathbf{A}}_s$ -driven VAG conductivity $\sigma_s^{x;\tilde{x}\tilde{x}}$ is obtained by replacing the vertex operator $\mathcal{V}_s^\alpha$ in the $\mathbf{A}_s$ -driven VAG conductivity $\sigma_s^{x;xx}$ with $\tilde{\mathcal{V}}_s^\alpha$.

Here, we focus on the NRSF terms because the contributions of the NRSF terms are dominant in the μeV -order frequency region, which is atypical frequency of the SAWs. The

type	$H_\xi^{\alpha\beta}(\mathbf{k})$	(α, β)	coupling to $\tilde{\mathbf{A}}_s$
A	$v_0\pi$	$(A_l, B_l) \quad (l = 1, 2, 3)$	No coupling
B	const.	$(B_1, A_2), (B_2, A_3), (A_1, B_3)$	No coupling
C1	$v_3\pi$	$(B_2, A_1), (B_3, A_2)$	$\mathbf{k} \rightarrow \mathbf{k} + r_3 \tilde{\mathbf{A}}_s$
C2	$v_4\pi$	$(A_1, A_2), (B_2, B_3)$	$\mathbf{k} \rightarrow \mathbf{k} + r_4 \tilde{\mathbf{A}}_s$

Table C.1: Off-diagonal matrix elements of the six-band model Hamiltonian $H_\xi^{\alpha\beta}(\mathbf{k})$ and couplings to the pseudogauge field $\tilde{\mathbf{A}}_s$. We ignore the spin degree of freedom.

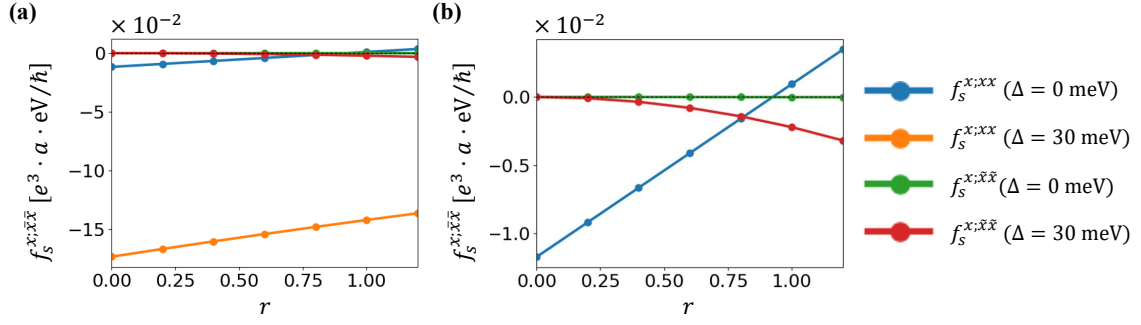


Figure C.1: The NRPSF $f_s^{x;xx}$ and $f_s^{x;\tilde{x}\tilde{x}}$ in the normal state ($\Delta = 0 \text{ meV}$) and IVC state ($\Delta = 30 \text{ meV}$). The r -range is $0 \leq r \leq 1.2$. Figure C.1(b) shows the enlarged view of Fig. C.1(a).

NRSF term of $\sigma_s^{x;\tilde{x}\tilde{x}}$ is given by

$$\sigma_{s,\text{NRSF}}^{x;\tilde{x}\tilde{x}} = -\frac{1}{2\Omega^2} \lim_{\lambda, \tilde{\lambda} \rightarrow 0} \partial_\alpha \partial_{\tilde{\beta}} \partial_{\tilde{\gamma}} F[\mathbf{A}_v = \boldsymbol{\lambda}, \tilde{\mathbf{A}}_s = \tilde{\boldsymbol{\lambda}}] = -\frac{1}{2\Omega^2} f_s^{x;\tilde{x}\tilde{x}}. \quad (\text{C.2})$$

where we introduce $f_s^{x;\tilde{x}\tilde{x}}$ by replacing the $\mathbf{A}_s$ -differentiation of $f_s^{x;xx}$ with the $\tilde{\mathbf{A}}_s$ -differentiation. Figure C.1 plots the NRPSF $f_s^{x;xx}$ and $f_s^{x;\tilde{x}\tilde{x}}$ in the normal state ($\Delta = 0 \text{ meV}$) and in the IVC state ($\Delta = 30 \text{ meV}$) with varying r . In both the normal and IVC states, $f_s^{x;\tilde{x}\tilde{x}}$ is much smaller than $f_s^{x;xx}$. Therefore, the $\tilde{\mathbf{A}}_s$ -driven VAG conductivity $\sigma_s^{x;\tilde{x}\tilde{x}}$ is finite but negligibly small. Thus, we ignore the contribution of the $\tilde{\mathbf{A}}_s$ -driven VAG effects in the main text.

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