Nonlocal optical conductivity of Fermi surface nesting materials

Xiamin Huang1,4*, Xiao Jiang2, Bing Huang2, and Zhou Li1,3,4*

1GBA Branch of Aerospace Information Research Institute, Chinese Academy of Sciences, Guangzhou 510700, China;
2Beijing Computational Science Research Center, Beijing 100193, China;
3University of Chinese Academy of Sciences, Beijing 100049, China;
4Guangdong Provincial Key Laboratory of Terahertz Quantum Electromagnetics, Guangzhou 510700, China

Received October 9, 2022; accepted November 10, 2022; published online March 6, 2023

We investigate the nonlocal optical conductivity of Fermi surface nesting materials that support charge density waves (CDWs) or spin density waves (SDWs). The nonlocal optical conductivity contains information on correlations in electron fluids, which could not be accessed by standard optical probes. A half-metal emerges from doping a CDW, and similarly, a spin-valley half-metal emerges from doping an SDW. Based on the parabolic band approximation, we find that the Drude peak is shifted to a higher frequency and splits into two peaks in the nonlocal optical conductivity. We attribute this to the two Fermi velocities in the half-metal or spin-valley half-metal states.

density functional theory, near-field spectroscopy, charge density wave, optical conductivity

PACS number(s): 71.15.Mb, 71.45.Lr, 78.20.Bh, 68.37.Uv

Citation: X. Huang, X. Jiang, B. Huang, and Z. Li, Nonlocal optical conductivity of Fermi surface nesting materials, Sci. China-Phys. Mech. Astron. 66, 247011 (2023), https://doi.org/10.1007/s11433-022-2035-7

1 Introduction

The scattering-type scanning near-field optical microscope (s-SNOM) is frequently used to probe the response pattern of a specific material (e.g., graphene) [1-3] driven by electromagnetic waves oscillating in time and space. The spatial dispersion in the response pattern is characterized by the nonlocal optical conductivity in close connection to the complicated structure of the Fermi surface and electron correlations [4]. These short-range effects are washed out by the standard far-field optical probe, which only measures the optical conductivity in the long wavelength limit. The tip radius $a$ is approximately 25 nm [1-3] for the atomic force microscope operating in the tapping mode of s-SNOM. Thus, the wave vector $q$ is in the order of $1/a \approx 0.04 \text{ nm}^{-1}$. The nonlocal optical conductivity is crucial for plasmons (collective oscillations of electrons). The imaginary part of the nonlocal optical conductivity determines the dispersion of plasmons, while the real part determines the damping rate of plasmons. Plasmonic antennas, lenses, and resonators are successful [5] at concentrating electromagnetic energy into the subwavelength and deep-subwavelength volume. Density-independent plasmons were found in nodal-line semimetals [6], with possible applications in terahertz-stable topological metamaterials. While a weak electron-phonon interaction (EPI) is one of the basic assumptions in the Bardeen-Cooper-Schrieffer (BCS) theory [7] for conventional superconductivity, a strong EPI leads to interesting phenomena, such as polarons [8,9] and charge density waves (CDWs) [10-13]. The interplay of the spin degree of freedom and strong EPI leads to spin polarons [14] and spin density waves (SDWs) [15,16].
One prevailing understanding of CDWs is the Peierls instability [17] in one dimensional (1D) and the Fermi surface nesting in any dimension. In many real materials, the Peierls instability fails, and the CDW phase is determined from the momentum dependence of the electron-phonon coupling matrix element [18]. Ref. [19] pointed out that a spin-valley half-metal state emerges from doing an SDW state. Then, ref. [20] clarified that a weak repulsive electron-electron interaction contributes to SDW ordering, following a BCS-like mean-field approach. The Fermi surface in the half-metal state is fully spin polarized. The nesting vector connecting particle and hole valleys gives inter-valley scattering. In principle, if no EPI is considered, the SDW order is always more energy favored than the CDW. In the presence of an EPI, one needs to consider the spin-flip process when the electrons are scattered by the lattice. The four-band Hamiltonian for the spin-valley half-metal has also been applied in twisted bilayer graphene [21, 22]. In two-dimensional (2D) valleytronic materials, such as MoS2 [23,24], two nonequivalent valleys are separated in the Brillouin zone by a large momentum, so the inter-valley scattering is very small. Manipulating spin has lead to spintronics [25,26]; similarly, manipulating the valley index can produce new effects, including using it to carry information. As an example, in the context of graphene, the ref. [27] showed that a contrasting intrinsic magnetic moment and Berry curvature are associated with the carrier valley index. The interband optical Hall conductivity [24] is then shown to be connected to the Berry curvature of the material, adding a dynamical factor to the static Berry curvature. The intraband longitudinal optical conductivity has been less investigated. In the spin-valley half-metal (doping-form SDW) or half-metal (doping-form CDW), the spin- and valley-polarized Fermi surface and Fermi velocities are, however, closely connected to the intraband nonlocal optical conductivity. Recently, in transition metal dichalcogenides (e.g., VSe2, MoTe2, and WTe2 and electron-doped MoS2 [28-31]) CDW phases were found. Half-metals are predicted in 2D metal selenides, such as V-doped SnSe2 and Co2Se3 [32]. In g-C3N4 and g-C4N3, whose electrons are entirely from the s- and p-orbitals, metal-free half-metals are predicted [33,34]. They may be used in biocompatible applications, e.g., in synergistic photothermal treatments to kill cancer cells [35]. In this work, we present the density functional theory (DFT) calculations of Fermi surface nesting candidate materials, 1T-VSe2 and g-C3N4, and provide an approximate tight binding Hamiltonian for 1T-VSe2. For the intraband nonlocal optical conductivity, the anisotropy of the band structure averages out, so we choose an isotropic parabolic band Hamiltonian to capture the important physics. We then use the Green’s function to calculate the nonlocal optical conductivity based on a velocity-velocity correlation function approach. Intra-band and interband optical conductivities are obtained in 1D, 2D, and three dimensions (3D). The nonlocal (q ≠ 0) optical conductivity becomes anisotropic in 2D and 3D, a feature not available in the (q = 0) optical conductivity. The imaginary part of the nonlocal optical conductivity shows two peaks in the intraband region for some parameters of the Fermi surface nesting model, suggesting two branches of plasmons in the intraband region.

2 Fermi surface nesting candidate materials

The numerical calculations on the mechanical and electronic properties of VSe2 are performed using a first-principles method based on the DFT [36], as implemented in the D- projector augmented wave (PAW), which is a program under the Device Studio platform. The DS-PAW is based on the plane-wave basis and the PAW representation [37]. The Perdew-Burke-Ernzerhof (PBE) exchange-correlation energy functional within the generalized gradient approximation is employed [38]. In the calculations, atomic positions are fully relaxed until the force on each atom is less than 0.05 eV/Å. The electronic iteration convergence criterion is set to 10−4 eV. The wave functions are expanded in plane waves up to a kinetic energy cutoff of 520 eV. The Brillouin zone integration is obtained using a k-point sampling mesh of 13 × 13 × 1, generated according to the Gamma-centered method. In the standard DFT calculations, which are used in this section, the staggered Hartree term representing the spin off-diagonal exchange energy is not included [39]. In the future, we plan to develop atomistic simulations for the SDW and CDW, similar to the DFT based on the Migdal-Eliashberg formalism [40].

In Figure 1(a), we present a schematic of the structure of 1T-VSe2 in the trigonal phase corresponding to the space group P3m1. For a single-layer 1T-VSe2, the primitive cell contains three atoms, the optimized lattice constant is a = b = 3.33 Å, and the angle between them is γ = 120°. In Figure 1(b), we show a super-cell of VSe2 and mark the three unit cells (000), (100), and (110). The black arrows represent the hopping between the nearest-neighbor atoms, including the Se1-Se2, V-Se1, and V-Se2 hoppings in the unit cell (000) and the V-V hopping between the unit cells (000) and (100). The blue arrows represent the second-nearest neighbor hoppings, including the Se1-Se4 and Se2-Se2 hoppings, between the unit cells (000) and (110). In Tables 1-3, we list the Wannier fitting results of the Se-Se and V-Se hopping parameters. The V-V hopping parameter is small, but the next-nearest Se-Se hopping parameter is large. The fitted tight binding model is a very large Hamiltonian matrix to be manipulated numerically. In Tables 1-3, we provide an approximate tight binding
model, as given in the Appendix. The anisotropy of the tight binding model averages out in the intraband conductivity, so we use the isotropic parabolic band approximation to capture the important physics of the many-body calculations in sect. 3.

![Figure 1](image)

**Figure 1** (Color online) Red balls represent V atoms; green balls represent Se atoms. A unit cell with one V atom and two Se atoms is marked in the bottom frame. (a) Crystal structure of 1T-VSe₂; (b) hopping between the nearest and next-nearest atoms.

| Table 1  | Se₁-Se₂ hopping amplitudes. |
|----------|-------------------------------|
|          | Se₂                          |
|          | pₓ  | pᵧ  | pₛ  | pₓ  | pᵧ  | pₛ  |
| Se₁      | 0.464 | -0.316 | 0.186 | -0.316 | 0.126 | -0.089 |
|          | 0.186 | -0.089 | 0.036 |

| Table 2  | V-Se₁ and V-Se₂ hopping amplitudes |
|----------|-----------------------------------|
|          | Se₁                            | Se₂                            |
|          | pₓ  | pᵧ  | pₛ  | pₓ  | pᵧ  | pₛ  | pₓ  | pᵧ  | pₛ  | pₓ  | pᵧ  | pₛ  |
|          | 0.515 | 0.481 | 0.283 | -0.518 | 0 | 0.568 |
|          | 0.487 | -0.750 | -0.729 | 0 | -0.500 | 0 |
| V        | 0.278 | -0.738 | 0.095 | 0.554 | 0 | 1.160 |
|          | -0.379 | -0.139 | 0.624 | -0.761 | 0 | -0.389 |
|          | 0.655 | 0.464 | -0.148 | 0 | -0.696 | 0 |

| Table 3  | Se-Se next nearest hopping amplitude |
|----------|-------------------------------------|
|          | pₓ₋pₓ  | pₓ₋pᵧ  | pₓ₋pₛ  | pᵧ₋pₓ  | pᵧ₋pᵧ  | pᵧ₋pₛ  |
|          | Se₁(000)>Se₂(110): -0.169 | 0.193 | 0.344 | 0.553 | 0.535 | 0.654 |
|          | Se₂(000)>Se₂(110): -0.169 | 0.193 | 0.553 | 0.344 | 0.654 |

In Figure 2, we show the band structure and Fermi surface of 1T-VSe₂ and g-C₄N₃. In Figure 2(a), the spin-polarized calculations predict that the monolayer 1T-VSe₂ is a magnetic metal, of which the magnitude of the magnetic moment is 0.6 μᵦ/cell (DFT result). The Wannier fitting of the DFT band structure is implemented within the Wannier90 code [41]. We construct the Hamiltonian in the Wannier basis using five d-orbitals of the V atom and six p-orbitals of two Se atoms to generate the localized Wannier functions. Overall, the Wannier-fitted band structure agrees well with the DFT one. Some experimental data from the angle-resolved photoemission spectroscopy of VSe₂ agree well with the non-spin-polarized DFT [42], whereas other experimental data of the Fermi surface agree well with the spin-polarized DFT [43], so we present both of them in Figure 2(a) and (b). For the bulk VSe₂, the band structure is similar to 2D VSe₂ near the Γ, M, and K points, as given in the Appendix. There are...
more high symmetry momentum points H, A, and L for the bulk VSe₂, and the band structure around these points is different as expected. In Figure 2(c), the spin-polarized calculations predict that g-C₄N₃ is a half-metal. The direct band gap for the spin-up electrons is 2.20 eV, and the magnetic moment of 1 μB/cell is found for g-C₄N₃. In the Fermi surface of g-C₄N₃, a nesting vector Q clearly connects the circle around the Γ point and K point. In all the calculations, the staggered Hartree term is not included. In the following sections, we use a parabolic band approximation to discuss the impact of the staggered Hartree term.

### 3 Hamiltonian and nonlocal optical conductivity

Below, we use the parabolic band approximation and discuss the many-body calculation for the nonlocal optical conductivity in the case of Fermi surface nesting in 1D, 2D, and 3D. With a nesting vector Q to connect (part of) the Fermi surface of a conduction band to that of a valence band, the Hamiltonian for a CDW state is given by

\[
\hat{H} = \begin{pmatrix}
\frac{k^2}{2m} - \varepsilon_0 & 0 & 0 & \Delta_1 \\
0 & \frac{k^2}{2m} - \varepsilon_0 & \Delta_1 & 0 \\
0 & \Delta_1 & \frac{k^2}{2m} + \varepsilon_0 & 0 \\
\Delta_1 & 0 & 0 & \frac{k^2}{2m} + \varepsilon_0
\end{pmatrix},
\]

where Δ₁ and Δ₂ are the gap parameters for a CDW insulator. In this study, we assume that the gap parameters are real numbers, so the Hamiltonian (eq. (1)) is a Hermitian matrix. In the case of complex gap parameters, two of Δ₁ and Δ₂ in eq. (1) should be replaced by Δ*₁ and Δ*₂ to keep it a Hermitian matrix. The basis states for the Hamiltonian are \[|c_{σk}⟩, |c_{σk}†⟩, |c_{σk+Q}⟩, |c_{σk+Q}†⟩\]; here, \[c_{σk} = \text{creation operator for a fermionic field with the wave vector k, spin } σ \]

and valley a. \[σ = ±1 \] is for the up and down directions of the spin. The SDW phase is connected to the CDW phase by transformations \[c_{hσ} → c_{hσ}†, c_{hσ}† → c_{hσ} \] with the corresponding SDW order parameter defined as \[\Delta_{σ} = (c_{σσ}c_{σσ}†)\] (here \(σ = ±σ\)). Although the SDW and CDW are connected mathematically, in the experiment, the detection is very different. X-ray diffraction and neutron scattering are used to detect the CDW, whereas only neutron scattering is used to detect the SDW. The gap parameters are generally determined by a variational principle based on the free energy of the system. For example, in ref. [19], it was found that \[Δ_λ(x) = Δ₀ + \sqrt{1 - x/(Δ₀N_F)}, Δ_μ(x) = Δ₀, \text{ and } μ = Δ₀ - x/2N_F \]

where \[x \text{ is the partial doping } x = -xΩ/Ω, Ω \text{ is the grand potential, and } μ \text{ is the chemical potential.} \] The Green’s function is defined as the inverse of \[z - \hat{H}, \text{ where } z = iω_ν \text{ for the imaginary frequency or } z = ω + i0^\text{−} \text{ for the real frequency:} \]

\[
\hat{G}(k, z) = \frac{1}{z - \hat{H}} = \begin{pmatrix}
G_{xx} & G_{ch} \\
G_{hc} & G_{hh}
\end{pmatrix}.
\]

We first define a velocity-velocity correlation function \[\Pi_{xx}(k, ω)\] which depends on the wave vector and imaginary frequency, \[\Pi_{xx}(q, iω_ν) = T \sum_{k} \text{Tr}(\hat{V}_v\hat{G}(k - q/2, iω_ν)\hat{V}_v\hat{G}(k + q/2, iω_ν + iω_ν)), \]

where \[ω_ν = 2πν/τ \text{ and } ω_ν = (2ν + 1)πT \text{ are the bosonic and fermionic Matsubara frequencies and } n \text{ and } l \text{ are the integers.} \]

The nonlocal optical conductivity in the real frequency is obtained from the Kubo formula by performing the analytical continuation \[iω_ν → ω + i0^\text{−}. \]

\[
σ_{xx}(q, ω) = -\frac{e^2}{iω} Π_{xx}(q, iω_ν → ω + i0^\text{−}).
\]

The matrix Green’s function \[\hat{G}(k, z)\] can be expanded in terms of a matrix spectral density \[\hat{A}(k, ω)\] as:

\[
\hat{G}(k, z) = \int_0^∞ \frac{dω}{2π} \hat{A}(k, ω), \]

The matrix spectral density function is proportional to the imaginary part of the matrix Green’s function:

\[
\hat{A}(k, ω) = 2\text{Im}[\hat{G}(k, ω)].
\]

After taking the sum over the imaginary frequencies \[iω_ν, \text{ the longitudinal conductivity } \ σ_{xx}(ω) \text{ is given by}

\[
σ_{xx}(q, ω) = -\frac{e^2}{4π^2} \int_0^∞ \frac{dω_1}{dω_2} F(ω) \times \sum_k \text{Tr}(\hat{V}_v\hat{A}(k - q/2, ω_1)\hat{V}_v\hat{A}(k + q/2, ω_2)),
\]

where \[F(ω) = |f(ω_1) - f(ω_2)|\] and \[f(x) \text{ is the Fermi-Dirac distribution function defined as } f(x) = 1/[\exp(x/T - μ/T) + 1]. \]

The real part of the optical conductivity can be obtained as:

\[
\text{Re}σ_{xx}(q, ω) = \frac{e^2}{4πω} \int_0^∞ \frac{dω_1}{dω} |f(ω_1) - f(ω_1 + ω)| \times \sum_k \text{Tr}(\hat{V}_v\hat{A}(k - q/2, ω_1)\hat{V}_v\hat{A}(k + q/2, ω_1 + ω)).
\]

The spectral function eq. (6) is a delta function if no impurity or other similar scatterings are considered. Here, we consider the self-energy correction from the impurity scattering to be a pure imaginary number, i.e., \[i/τ \]. The spectral function eq. (6) is then a broadened delta function, and the integral over \[ω_1 \]

\[...\]
in eq. (8) could not be removed. From eqs. (7) and (8), we write the imaginary part of the optical conductivity as:
\[ \text{Im} \sigma_{xx}(q, \omega) = \frac{1}{\pi} \int_{-\infty}^{\infty} \text{Re} \sigma_{xx}(q, \omega') \frac{d\omega'}{\omega - \omega'} , \]
(9)
which recovers the Kramers-Kronig relations.

In 1D, the corresponding velocity operator can be obtained as (for simplicity, we set \( h = 1 \)):
\[ \hat{v}_x(k) = \frac{\partial H}{\partial k} = \begin{pmatrix} (k/m)I_2 & 0 \\ 0 & -(k/m)I_2 \end{pmatrix} , \]
(10)
where \( I_2 \) is the \( 2 \times 2 \) unit matrix. For the CDW state, the spin operator is changed to \( \hat{S}_z = \text{diag}(-1, 1, 1, -1) \). For the SDW state, the spin-valley operator is \( \tau_z = \text{diag}(1, 1, 1, 1) \), while the spin-valley operator for the SDW is the same as the spin-valley operator for the CDW. For both cases, the valley operator is \( \tau_x = \text{diag}(1, 1, 1, 1) \). With these operators, we define the spin conductivity (for a CDW), spin-valley conductivity (for a SDW), and valley conductivity (for both). Details are given in the Appendix.

4 Results and discussion

In Figure 3, we present the numerical results of the optical conductivity based on the DFT calculations in sect. 2. In the DFT software (DS-PAW) we use, the real part of the optical conductivity is calculated from the imaginary part of the dielectric function. The DFT calculations do not catch the intraband part of the nonlocal optical conductivity, usually in the frequency region from 0 to 50 meV. Moreover, in the standard DFT calculation, the many-body effect from the Fermi surface nesting is missing because the staggered Hartree term is not included. Below, we present the numerical results of the nonlocal optical conductivity based on the Hamiltonian (eq. (1)) in sect. 3. The eigenvalues of the Hamiltonian (eq. (1)) are obtained as \( E = \pm \sqrt{(k^2/m - \epsilon_0)^2 + \Delta^2} \), where \( \Delta = \Delta_1 \) or \( \Delta_0 \). We choose a typical wave vector \( k_0 = 1.0 \text{ nm}^{-1} \), whose corresponding energy is \( \epsilon_0 = 0.2E_0 \) and define \( x' = x/(N_F\Delta_0) \) in all the numerical simulations. The impurity scattering self-energy correction is \( 1/\tau = 0.005E_0 \). The parameters of the energy shift \( \epsilon_0 \), gap \( (\Delta_0, \Delta_1) \), and chemical potential \( \mu \) are determined from a variational principle calculation [19]. As suggested, the gap \( \Delta_0 \) is much smaller than \( \epsilon_0 \), (e.g., \( \Delta_0 = \epsilon_0/4 \)). Here, we choose \( \Delta_0 = \epsilon_0 \). For the intraband optical conductivity, tuning the band gap to be much smaller will not change the key feature, in the sense that the chemical potential still intersects the lower conduction band, and two Fermi velocities are found.

In Figure 4, we show a typical 1D band structure (left) or a 3D iso-energy surface (right) for the CDW/SDW phase. The doping parameter is \( x' = 0.9 \), so the gap parameters are \( \Delta_1 = 0.063E_0 \) and \( \Delta_1 = 0.2E_0 \). The doping \( x \) gives an asymmetry between the gap order parameter for the spin-up and spin-down, so the Fermi surface is now spin-polarized, typical for a half-metal. The Fermi velocities are depicted by arrows in the right panel. In 1D, \( q \) is a number, so we only have the longitudinal conductivity. In 2D and 3D, \( q \) is a vector, so we need to consider the longitudinal one \( (q_0 \text{ along } \hat{e}_z) \) and transverse one \([q \text{ perpendicular to } \hat{e}_z] \). For the longitudinal conductivity \( \sigma^{xx}(q, \omega) \), we have the absolute value of \( k + q = \sqrt{(k_x + q_x)^2 + k_z^2} = \sqrt{k^2 + 2kq\cos(\theta) + q^2} \). For the transverse one \( \sigma^{yy}(q, \omega) \), we have the absolute value of \( k + q = \sqrt{(k_x + q_x)^2 + k_z^2} = \sqrt{k^2 + 2kq\sin(\theta) + q^2} \). Because of this difference, we see a large anisotropy in the nonlocal optical conductivity in 2D and 3D.

In 3D, we need to numerically perform the fourfold integration (eq. (8)), which is very slow. One frequency point takes about 1 h on one CPU. In Figure 5(a), we use 1600 points and show the real part of the nonlocal optical conductivity in 3D for \( q = 0.2 \text{ nm}^{-1} \). We found that the Drude peak at the zero frequency is shifted to higher frequencies for the longitudinal conductivity. In the intraband region \((0 < \omega/E_0 < 0.2)\), the Drude peak is separated into a

![Figure 3](image-url)
Figure 4 (Color online) (a) Band structure for a Fermi surface nesting model in 1D with the SDW order parameters $\Delta_1 = 0.063E_0$ and $\Delta_2 = 0.2E_0$. The chemical potential $\mu = 0.11E_0$ is denoted with the red dashed line. (b) Schematics of the iso-energy surface of a Fermi surface nesting model in 3D, with two Fermi spheres assigned with two Fermi velocities. When the chemical potential is tuned, the two Fermi spheres move close to or far away from each other until one of them disappears. The Fermi velocities point outward with the same amplitude on one Fermi sphere and point inward with a smaller amplitude on the other, denoted by the black arrows.

Figure 5 (Color online) (a) Real part of the nonlocal optical conductivity for a 3D vector $\mathbf{q}$. The black solid curve represents the longitudinal conductivity $\sigma_{xx}^L(q,\omega)$ ($\mathbf{q}$ along $\hat{e}_x$), and the Drude peak shifts to higher energy and splits into two peaks in the intraband due to the two different Fermi velocities. The red dashed curve represents the transverse conductivity $\sigma_{xx}^T(q,\omega)$ ($\mathbf{q}$ perpendicular to $\hat{e}_x$), and the Drude peak is not shifted. (b) 2D vector $\mathbf{q}$ is considered. We observe similar behavior as those in the 3D vector. The second peak in the intraband is sharper than that in the 3D case. (c) Real part of the optical conductivity (black solid) for a doped CDW half-metal in 1D ($\mathbf{q} = (0.2, 0)$ nm$^{-1}$) and the spin optical conductivity (red dashed) compared with the valley optical conductivity (green dash-dotted). Clearly, the two intraband peaks are associated with the same direction of the spin but with an opposite valley. The two interband peaks are, however, associated with the opposite spin. (d) Imaginary part of the nonlocal optical conductivity, which is crucial for plasmonics; two intraband peaks in the imaginary part are confirmed for $\mathbf{q} = (0.2, 0)$ nm$^{-1}$ (red dashed), as found in Re$\sigma(q,\omega)$. In all the plots, the parameters ($\Delta_1 = 0.063E_0$, $\Delta_2 = 0.2E_0$) are the same for 1D, 2D, and 3D. The chemical potential is $\mu = 0.11E_0$ intersecting with the lower conduction band. The temperature is $T = 0.001$ K. In all the plots of Re$\sigma(q,\omega)$, the first two peaks (or one peak and one dip) in the region $0 < \omega/E_0 < 0.2$ are intraband peaks, originating from two Fermi velocities. Then, in the region $0.2 < \omega/E_0$, two interband peaks originate from the peaks in the joint density of states (DOSs) for Re$\sigma(q = 0, \omega)$ [46] or the joint spectral function for Re$\sigma(q \neq 0, \omega)$.

Two-peak structure, which is due to the two Fermi velocities in the CDW, as shown in Figure 4. If the two typical Fermi velocities are $v_{F1}$ and $v_{F2}$, the two intraband peaks are roughly at $\omega_1 = q \times v_{F1}$ and $\omega_2 = q \times v_{F2}$. For the transverse direction ($\mathbf{q}$ perpendicular to $\hat{e}_x$), the Drude peak is not shifted. In both cases, we found two interband peaks at approximately 0.2 and 0.4 because, for a four-band system, we can define two sets of interband transitions. The
interband peaks are shifted to higher energy as the transverse mode crosses over to the longitudinal mode. In Figure 5(b), we investigate the 2D conductivity and find that the spectral weight is redistributed between the intraband and interband peaks. The intraband peaks in 2D become sharper than those in 3D. In Figure 5(c), the 1D intraband spin conductivity is always positive, whereas the intraband valley conductivity oscillates from negative to positive. This finding suggests that the charge carriers at the two Fermi momenta carry the same spin and the opposite valley. Figure 5(d) shows the 1D imaginary part of the nonlocal optical conductivity, which is connected to the real part through the Kramers-Kronig relation. At $q = 0$, the imaginary part does not diverge as $1/\omega$, so $\omega \Im \sigma = 0$. At $q = 0.2 \text{ nm}^{-1}$, we clearly see that the intraband evolves into two positive oscillations. At much larger $q = 0.4 \text{ nm}^{-1}$, however, we only see one positive peak in the intraband. This finding suggests that at a small $q$, two plasmonic modes may be observed in the electron energy loss spectroscopy. The interband transitions in the spin susceptibility of the spin-valley half-metal were studied in ref. [45].

The interband optical conductivity is connected to the joint DOSs for $\Re \sigma(q = 0, \omega)$ [46] or the joint spectral function for $\Re \sigma(q \neq 0, \omega)$. In Figure 6, we plot the real part of the nonlocal optical conductivity in 1D. When the Drude peak at $q = 0$ is shifted to higher energy, the spectral weight is transferred to the intraband region. In the intraband, the Drude peak splits into two peaks due to the two different Fermi velocities. The interband peaks at $\omega/E_0 = 0.4$ are not changed much by $q$. The parameters are the same as those in Figure 5. In Figure 7, we investigate the off-diagonal conductivity $\sigma_{xy}(q, \omega)$, which is zero in the direction $\theta = 0$ and $\theta = \pi/2$. Here, we set the direction $\theta = \pi/4$ (between $q$ and $\hat{e}_x$). We show that $\sigma_{xy}$ at $\omega = 0$ is negative and oscillates between positive and negative values at higher frequencies. The diagonal conductivity $\sigma_{xx}$ is in the transition from the transverse ($\theta = \pi/2$) to the longitudinal ($\theta = 0$) mode. We also see a shifted Drude peak and a dip in the optical conductivity around $\omega/E_0 = 0.1$ due to the two Fermi velocities. In Figure 8, $\sigma_{xx}(q, \omega)$ oscillates as the angle changes from $\theta = 0$ to $\theta = 2\pi$ for four typical frequencies in the intraband region. For the three of them, $\sigma_{xx}(q, \omega)$ reaches maximum at $\theta = 0$, for one frequency at $\omega/E_0 = 0.05$, and the maximum is at $\theta = \pi/2$. In Table 4, we present the DC off-diagonal conductivity along the direction $\theta = \pi/4$ for various wave vectors $q$. We found that $\Im \sigma_{xx}(q, 0)$ is positive at a small $q$; it first increases, then decreases, and changes sign at a larger $q$. In the Appendix, we present the numerical results of 1D optical conductivity and discuss the spin and valley conductivity.

In the Fermi surface nesting model, frequent electron-electron collisions modify the band structure, and two Fermi velocities appear. In a hydrodynamic approach, considering...
a fluid flow, the velocity gradually changes from the center to the boundary. In this sense, the CDW/SDW state lies somewhere between the ballisitic transport picture and the fully hydrodynamic transport picture. The intraband optical conductivity, which could be measured in a near-field experiment, is a good probe of these liquid-like properties. Moreover, the valley conductivity for one intraband peak is negative, and that for another intraband peak is positive. This condition presents a clear definition of the valley. The positive valley is associated with the Fermi velocity $v_{F1}$, and the negative valley is associated with $v_{F2}$. In the above discussion, we found that the nonlocal intraband optical conductivity is a useful tool to investigate the spin-valley physics.

5 Conclusion

In summary, we provide DFT calculations of Fermi surface nesting materials. Then, based on the parabolic band approximation, we derive the nonlocal optical conductivity for a SDW/CDW Hamiltonian beyond the standard DFT. Our method can accurately capture the shift of the Drude peak from zero to higher frequencies in the intraband region and its splitting into two peaks, in 3D, 2D, and 1D. As the direction changes from the transverse mode ($\theta = \pi/2$) to the longitudinal mode ($\theta = 0$), although the Hamiltonian is isotropic, the nonlocal optical conductivity is anisotropic at nonzero $q$.

We discuss in detail the diagonal $\sigma_{xx}$ and off-diagonal $\sigma_{xy}$ conductivities at different angles.

A part of this work has been supported by the ISSP International Collaboration Program of the University of Tokyo. Zhou Li acknowledges the support of a JSPS Foreign Postdoctoral Fellowship (Grant Nos. PE14052, and P16027), and the Chinese Academy of Sciences funding (Grant Nos. E121D10200, and E222D10200). This work was supported by the National Natural Science Foundation of China (Grant No. 61988102), the Key Research and Development Program of Guangdong Province (Grant No. 2019B090917007), the Science and Technology Planning Project of Guangdong Province (Grant No. 2019B090906011), and the ZJ project (Grant No. 2021Q002X159). We gratefully acknowledge HZWTECH for providing computation facilities. The authors thank A. Rozhkov, A. Shoybchakov, A. L. Rakhmanov, K. I. Kugel, R. Matsunaga, and F. Nori for useful discussions.

Table 4 $\Im \sigma_{xy}(q, \omega = 0)$ along the direction $\theta = \pi/4$

| $q$    | $\Im \sigma_{xy}(q, 0)$    |
|--------|-----------------------------|
| 0.001  | 0.00025                     |
| 0.005  | 0.0745                      |
| 0.01   | -4.06                       |
| 0.05   | -17.5                       |

1 Z. Fei, G. O. Andreev, W. Bao, L. M. Zhang, A. S. McLeod, C. Wang, M. K. Stewart, Z. Zhao, G. Dominguez, M. Thiemens, M. M. Fogler, M. J. Tauber, A. H. Castro-Neto, C. N. Lau, F. Keilmann, and D. N. Basov, Nano Lett. 11, 4701 (2011), arXiv: 1112.0390.

2 Z. Fei, A. S. Rodin, G. O. Andreev, W. Bao, A. S. McLeod, M. Wagner, L. M. Zhang, Z. Zhao, M. Thiemens, G. Dominguez, M. M. Fogler, A. H. C. Neto, C. N. Lau, F. Keilmann, and D. N. Basov, Nature 487, 82 (2012), arXiv: 1202.4993.

3 M. B. Lundeberg, Y. Gao, R. Asgari, C. Tan, B. Van Dunphen, M. Au-tore, P. Alonso-González, A. Woesener, K. Watanabe, T. Taniguchi, R. Hillenbrand, J. Hone, M. Polini, and F. H. L. Koppens, Science 357, 187 (2017), arXiv: 1704.05518.

4 G. Giuliani, and G. Vignale, Quantum Theory of the Electron Liquid (Cambridge University Press, Cambridge, 2005).

5 J. A. Schuller, E. S. Barnard, W. Cai, Y. C. Jun, J. S. White, and M. L. Brongersma, Nat. Mater. 9, 193 (2010).

6 J. Wang, X. Sui, W. Duan, F. Liu, and B. Huang, Proc. Natl. Acad. Sci. USA 118, e2023029118 (2021), arXiv: 2010.11436.

7 J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. 106, 162 (1957).

8 Z. Li, C. J. Chandler, and F. Marsiglio, Phys. Rev. B 83, 045104 (2011), arXiv:1011.1259.

9 Z. Li, and F. Marsiglio, J. Supercond. Nov. Magn. 25, 1313 (2012).

10 G. Grüner, Rev. Mod. Phys. 60, 1129 (1988).

11 P. Monceau, Adv. Mat. 61, 325 (2012), arXiv: 1307.0929.

12 F. Tang, Y. Ren, P. Wang, R. Zhong, J. Schneeleoch, S. A. Yang, K. Yang, P. A. Lee, G. Gu, Z. Qiao, and L. Zhang, Nature 569, 537 (2019), arXiv: 1807.02678.

13 P. L. Zhao, H. Z. Lu, and X. C. Xie, Phys. Rev. Lett. 127, 046602 (2021), arXiv: 2012.12541.

14 Z. Li, L. Covaci, M. Berceu, D. Baille, and F. Marsiglio, Phys. Rev. B 83, 195104 (2011), arXiv: 1102.1421.

15 A. W. Overhauser, Phys. Rev. 128, 1437 (1962).

16 E. Fawcett, Rev. Mod. Phys. 60, 209 (1988).

17 R. Peierls, Ann. Phys. 396, 121 (1930).

18 X. Zhu, Y. Cao, J. Zhang, E. W. Plummer, and J. Guo, Proc. Natl. Acad. Sci. USA 112, 2367 (2015), arXiv: 1503.01858.

19 A. V. Rozhkov, A. L. Rakhmanov, A. O. Shoybchakov, K. I. Kugel, and F. Nori, Phys. Rev. Lett. 119, 107601 (2017), arXiv: 1704.01727.

20 A. L. Rakhmanov, A. O. Shoybchakov, K. I. Kugel, A. V. Rozhkov, and F. Nori, Phys. Rev. B 98, 155141 (2018), arXiv: 1811.04202.

21 C. J. Tabert, and E. J. Nicol, Phys. Rev. B 87, 121402 (2013), arXiv: 1302.7233.

22 A. O. Shoybchakov, A. V. Rozhkov, A. L. Rakhmanov, and F. Nori, Phys. Rev. Lett. 120, 266402 (2018), arXiv: 1707.08886.

23 D. Xiao, G. B. Liu, W. Feng, X. Xu, and W. Yao, Phys. Rev. Lett. 108, 196802 (2012), arXiv: 1112.3144.

24 Z. Li, and J. P. Carbotte, Phys. Rev. B 86, 205425 (2012), arXiv: 1211.3094.

25 S. A. Wolf, D. D. Awschalom, R. A. Buhrman, J. M. Daughton, S. von Molnar, M. L. Roukes, A. Y. Chtchelkanova, and D. M. Treger, Science 294, 1488 (2001).

26 J. Fabian, A. Matos-Abiague, C. Ertler, P. Stano, and I. Zutic, Acta Phys. Slovaca 57, 565 (2007).

27 D. Xiao, W. Yao, and Q. Niu, Phys. Rev. Lett. 99, 236608 (2007), arXiv: 0709.1274.

28 J. Feng, D. Biswas, A. Rajan, M. D. Watson, F. Mazzola, O. J. Clark, K. Underwood, I. Markovic, M. McLaren, A. Hunter, D. M. Burn, L. B. Duffy, S. Barua, G. Balakrishnan, F. Bertran, P. Le Fèvre, T. K. Kim, G. van der Laan, T. Hesjedal, P. Wahl, and P. D. C. King, Nano Lett. 18, 4493 (2018).

29 J. Diego, A. H. Said, S. K. Mahatha, R. Bianco, L. Monacelli, M. Calandra, F. Mauri, K. Rossnagel, I. Errea, and S. Blanco-Canosa, Nat. Commun. 12, 598 (2021).

30 G. Marini, and M. Calandra, Phys. Rev. Lett. 127, 257401 (2021), arXiv: 2111.11920.

31 M. K. Bin Shubhan, A. Suleman, G. Moore, P. Phu, M. Hoesch, H. Kurebayashi, C. A. Howard, and S. R. Schofield, Nano Lett. 21, 5516 (2021), arXiv: 2108.07015.

32 C. Tang, L. Zhang, Y. Jiao, C. Zhang, S. Sanvito, and A. Du, J. Mater. Chem. C 9, 12046 (2021).
23 A. Du, S. Sanvito, and S. C. Smith, Phys. Rev. Lett. 108, 197207 (2012).
24 I. Choudhuri, G. Bhattacharyya, S. Kumar, and B. Pathak, J. Mater. Chem. C 4, 11530 (2016).
25 J. Yang, R. Xie, L. Feng, B. Liu, R. Lv, C. Li, S. Gai, F. He, P. Yang, and J. Lin, ACS Nano 13, 13144 (2019).
26 G. Kresse, and D. Joubert, Phys. Rev. B 59, 1758 (1999).
27 P. E. Blochl, Phys. Rev. B 50, 17953 (1994).
28 J. P. Perdew, K. Burke, and M. Ernzerhof, Phys. Rev. Lett. 77, 3865 (1996).
29 K. Capelle, M. F. Silva, and L. N. Oliveira, J. Magn. Magn. Mater. 226-230, 1017 (2001).
30 D. Wu, Y. Lin, L. Xiong, J. Li, T. Luo, D. Chen, and F. Zheng, Phys. Rev. B 103, 224502 (2021), arXiv: 2105.14243.
31 N. Marzari, A. A. Mostofi, J. R. Yates, I. Souza, and D. Vanderbilt, Rev. Mod. Phys. 84, 1419 (2012), arXiv: 1112.5411.
32 P. Chen, W. W. Pai, Y. H. Chan, V. Madhavan, Y. M. Chou, S. K. Mo, A. V. Fedorov, and T. C. Chang, Phys. Rev. Lett. 121, 196402 (2018), arXiv: 1811.05690.
33 G. Durja, B. K. Choi, T. Thi Ly, N. H. Lam, K. Jang, D. D. Dung, Y. J. Chang, and J. Kim, Nanotechnology 32, 364002 (2021).
34 P. Rodriguez-Lopez, W. J. M. Kort-Kamp, D. A. R. Dalvit, and L. M. Woods, Phys. Rev. Mater. 2, 014003 (2018), arXiv: 1711.06311.
35 D. A. Khokhlov, A. L. Rakhamonov, A. V. Rozhkov, and A. O. Shyvakov, Phys. Rev. B 101, 235141 (2020), arXiv: 2002.05504.
36 Z. Li, F. Marsiglio, and J. P. Carbotte, Sci. Rep. 3, 02828 (2013), arXiv: 1310.3916.

Appendix

A1 Derivations of the Green’s function and the spectral function for a four-band Hamiltonian

To obtain the Green’s function associated with the Hamiltonian (1), we first use a general mathematical technique: any 4 × 4 matrix can be partitioned into a smaller 2 × 2 block matrix. In the smaller block matrix, each element is a 2 × 2 sub-matrix. Using A, B, C, D for the four sub-matrices, we have

\[ z - \hat{H} = \begin{bmatrix} A & C \\ D & B \end{bmatrix}, \]  

(\text{a1})

where \( z = \text{i} \omega_n \) for the imaginary frequency or \( z = \omega + \text{i} \delta \) for the real frequency. Here, \( A = (z - k^2/2m + \epsilon_0)I_2 \), \( B = (z + k^2/2m - \epsilon_0)I_2 \), and

\[ C = \begin{bmatrix} 0 & -\Delta_1 \\ -\Delta_\uparrow & 0 \end{bmatrix}, \quad D = \begin{bmatrix} 0 & -\Delta_\downarrow \\ -\Delta_\downarrow & 0 \end{bmatrix}. \]  

(\text{a2})

The Green’s function matrix is also partitioned into a smaller 2 × 2 block matrix, with \( G_{ee}, G_{eh}, G_{he}, G_{hh} \) as the four sub-matrix elements. The equation to determine the Green’s function is written as:

\[ \begin{bmatrix} A & C \\ D & B \end{bmatrix} \begin{bmatrix} G_{ee} & G_{eh} \\ G_{he} & G_{hh} \end{bmatrix} = I_2 \begin{bmatrix} 0 & \hat{I}_2 \\ \hat{I}_2 & 0 \end{bmatrix}. \]  

(\text{a3})

The sub-matrix elements \( G_{ee} \) and \( G_{he} \) satisfy the following equations:

\[ A \times G_{ee} + C \times G_{he} = \hat{I}_2, \]  

(\text{a4})

\[ B \times G_{he} + D \times G_{ee} = 0. \]  

(\text{a5})

From the two equations, we obtain

\[(AB - CD)G_{ee} = B, \]  

(\text{a6})

\[(AB - DC)G_{he} = -D. \]  

(\text{a7})

The product of \( CD \) is given by

\[ \begin{bmatrix} 0 & -\Delta_\uparrow \\ -\Delta_\uparrow & 0 \end{bmatrix} \begin{bmatrix} 0 & \Delta_\downarrow \\ \Delta_\downarrow & 0 \end{bmatrix} = \begin{bmatrix} \Delta_\downarrow^2 & 0 \\ 0 & \Delta_\downarrow^2 \end{bmatrix}. \]  

(\text{a8})

For the other two sub-matrix elements \( G_{eh} \) and \( G_{hh} \), we need to solve the two equations below:

\[ A \times G_{eh} + C \times G_{hh} = 0, \]  

(\text{a9})

\[ D \times G_{eh} + B \times G_{hh} = \hat{I}_2, \]  

(\text{a10})

from which we obtained

\[ (AB - CD)G_{eh} = -C, \]  

(\text{a11})

\[ (AB - DC)G_{hh} = A. \]  

(\text{a12})

The poles of the four Green’s functions are given by the following equation:

\[ (z - k^2/2m + \epsilon_0)(z + k^2/2m - \epsilon_0) - \Delta^2 = 0, \]  

(\text{a13})

where \( \Delta = \Delta_\uparrow \) or \( \Delta_\downarrow \). Expand this equation in orders of \( z \), and we have

\[ z^2 - (k^2/2m - \epsilon_0)^2 - \Delta^2 = 0. \]  

(\text{a14})

Accordingly, we obtain two groups of solutions for the eigenvalues corresponding to \( \Delta = \Delta_\uparrow \) and \( \Delta_\downarrow \) separately:

\[ E_{1,2} = \pm \sqrt{\frac{k^2}{2m} - \epsilon_0}^2 + \Delta_\uparrow^2, \]  

(\text{a15})

and the other two for \( \Delta_\downarrow \),

\[ E_{3,4} = \pm \sqrt{\frac{k^2}{2m} - \epsilon_0}^2 + \Delta_\downarrow^2. \]  

(\text{a16})

With the eigenvalues \( E_{1,2,3,4} \), we can write the Green’s function in a compact form:

\[ G_{ee} = \begin{bmatrix} (z + k^2/2m - \epsilon_0) & 0 \\ -E_2(z - E_4) & (z + k^2/2m - \epsilon_0) \end{bmatrix}, \]  

(\text{a17})

\[ G_{eh} = \begin{bmatrix} 0 & \Delta_\uparrow \\ (z - E_4)(z - E_2) & 0 \end{bmatrix}, \]  

(\text{a18})
To evaluate the conductivity, we need to take the trace of the nary part of the Green’s function, we need to use the following rules to decompose the Green’s function into a sum of single poles:
\[
\frac{(z + k^2/2m + \epsilon_0)}{(z - E_1)(z - E_2)} = \frac{1}{(E_1 - E_2)} \left[ \frac{E_1 + k^2/2m + \epsilon_0}{(z - E_1)} - \frac{E_2 + k^2/2m + \epsilon_0}{(z - E_2)} \right],
\]
\[
\frac{(z - k^2/2m + \epsilon_0)}{(z - E_1)(z - E_2)} = \frac{\Delta_t}{(E_1 - E_2)} \left[ \frac{E_1 - k^2/2m + \epsilon_0}{(z - E_1)} - \frac{E_2 - k^2/2m + \epsilon_0}{(z - E_2)} \right].
\]

The DOS \( N(\omega) \) can be obtained through the spectral function: \( N(\omega) = \sum_k \text{Tr}[\hat{A}(k, \omega)] \).

### A2 Derivation of the nonlocal optical conductivity

To evaluate the conductivity, we need to take the trace of the matrix product of the velocity operator and Green’s function matrix. For \( \sigma^\parallel_{z\omega}(\omega) \), we have
\[
\text{Tr}(\hat{v}_z \hat{G}(k, \omega) \hat{v}_z \hat{G}(k, \omega)) = (k_z/m)^2 
\times \left[ \text{Tr}[G_{ee}(k, \omega) G_{ee}(k, \omega)] - \text{Tr}[G_{eh}(k, \omega_1) G_{he}(k, \omega_2) + G_{he}(k, \omega_1) G_{eh}(k, \omega_2)] + \text{Tr}[G_{hh}(k, \omega_1) G_{hh}(k, \omega_2)] \right].
\]

The spin and valley conductivities are given as:
\[
\text{Re} \sigma^z_{xx}(q, \omega) = \frac{e^2}{4m_0} \int_0^\infty d\omega_1 [f(\omega_1) - f(\omega_1 + \omega)] \times 
\sum_k \text{Tr}(S_z \hat{v}_z \hat{A}(k - q/2, \omega_1) \hat{v}_z \hat{A}(k + q/2, \omega_1 + \omega))
\]
and
\[
\sum_k \text{Tr}(S_z \hat{v}_z \hat{A}(k - q/2, \omega_1) \hat{v}_z \hat{A}(k + q/2, \omega_1 + \omega)),
\]
respectively. For the CDW, we define the spin conductivity \( \sigma^z_{xx}(\omega) \). The spin operator is \( S_z = \text{diag}(-1, 1, 1, -1) \); for the SDW, the spin operator becomes \( S_z = \text{diag}(-1, 1, 1, -1) \); the spin valley operator is \( S_z \tau_z = \text{diag}(-1, 1, 1, -1) \), the same as the spin operator for the CDW. Mathematically, for the spin conductivity \( \sigma^z_{xx}(\omega) \) (CDW) and spin valley conductivity \( \sigma^z_{yy}(\omega) \) (SDW), we need the same trace, i.e.,
\[
\begin{align*}
&\text{Tr}(S_z \hat{v}_z \hat{G}(k, \omega_1) \hat{v}_z \hat{G}(k, \omega_2)) = (k_z/m)^2 
\times \left[ \text{Tr}[\sigma \hat{G}_{ee}(k, \omega_1) \hat{G}_{ee}(k, \omega_2)] - \text{Tr}[\sigma \hat{G}_{he}(k, \omega_1) \hat{G}_{he}(k, \omega_2) + \hat{G}_{he}(k, \omega_1) \hat{G}_{he}(k, \omega_2)] \right] \\
&\text{Tr}[\sigma \hat{G}_{hh}(k, \omega_1) \hat{G}_{hh}(k, \omega_2)]].
\end{align*}
\]

For the valley conductivity \( \sigma^y_{xx}(\omega) \), we need
\[
\begin{align*}
&\text{Tr}(\tau_z \hat{v}_z \hat{G}(k, \omega_1) \hat{v}_z \hat{G}(k, \omega_2)) = (k_z/m)^2 
\times \left[ \text{Tr}[\hat{G}_{ee}(k, \omega_1) \hat{G}_{ee}(k, \omega_2)] - \text{Tr}[\hat{G}_{he}(k, \omega_1) \hat{G}_{he}(k, \omega_2) + \hat{G}_{he}(k, \omega_1) \hat{G}_{he}(k, \omega_2)] \right] \\
&\text{Tr}[\hat{G}_{hh}(k, \omega_1) \hat{G}_{hh}(k, \omega_2)].
\end{align*}
\]

In the clean limit where the impurity scattering self-energy vanishes, the spectral function reduces to \( \delta \)-function; for example, \( A_{xx}(k, \omega) = \frac{-2\pi}{E_x' \delta(\omega - E_x) - E_x \delta(\omega - E_x)} \frac{0}{(E_x - E_x') \delta(\omega - E_x)} \frac{0}{(E_x' - E_x) \delta(\omega - E_x')} \).

Here, \( E_x = E_n + k^2/2m - \epsilon_0 \) and \( n = 1, 2, 3, 4 \). The trace \( \text{Tr}[\hat{G}_{ee}(k, \omega_1) \hat{G}_{ee}(k, \omega_2)] = \frac{1}{(E_n - E_2)^2} \). In 1D, the nonlocal optical conductivity contains the term \( \frac{1}{E_{\text{Drude}}(k - q/2)E_{\text{Drude}}(k + q/2)} \), where \( E_{\text{Drude}}(k, \omega_1, \uparrow) = \frac{k_z^2/2m - \epsilon_0}{(E_n - E_2)(E_n - E_1)} \) and \( E_{\text{Drude}}(k, \omega_1, \downarrow) = \frac{k_z^2/2m - \epsilon_0}{(E_n - E_2)(E_n - E_1)} \). The two \( \delta \)-functions could be used to perform the integral over \( \omega_1 \), and we are left with the 1D integral over \( k \). The upper and lower limits in the integral over \( k \) are set by the Fermi-Dirac distribution function. The analytical results will be very long and complicated because many terms from the intraband and interband need to be considered. Instead, we decide to perform the integration numerically. The self-energy from impurity scattering is nonzero in all the numerical results.

In Figure a1, we show the real part of the optical conductivity for various wave vectors \( q \). For \( q = 0 \), we recover the famous Drude peak for the intraband optical conductivity. For the nonzero wave vector \( q \), we observe the shift of the Drude peak to higher frequencies. The single Drude peak is divided into two peaks, which are associated with the two typical energies for the intraband transition. This is due to the fact that in 1D, the slope near the four Fermi momenta (the momenta at which the Fermi surface intersects with the band) are different. The slope is related to the Fermi velocity. There are two typical Fermi velocities \( v_{F1} \) and \( v_{F2} \). Thus, for
parameters are given in Table 2 of the main text. The model represents the hopping between the V and Se atoms. The hopping to describe the hoppings on the lattice. The solid line represents the hopping between the Se1 and Se2 atoms. The hopping parameters are given in Table 2 of the main text. Here, $\delta_1 = (1.667, 0.962), \delta_2 = (0.000, -1.924)$, and $\delta_3 = (-1.667, 0.962)$ in the unit of Å. The tight binding

![Figure a1](image1.png)

**Figure a1** (Color online) Real part of the optical conductivity for $\Delta_1 = 0.063E_0$ and $\Delta_1 = 0.2E_0$ and for wave vectors $q = 0$, $q = 0.2$ nm$^{-1}$, and $q = 0.4$ nm$^{-1}$. The chemical potential $\mu = 0.11E_0$. For $q = 0$ (black solid), a Drude peak from the intraband transition has a near-zero frequency. At higher frequencies $\omega = 0.2E_0$ and $\omega = 0.4E_0$, two peaks from the interband transition are found due to the four-band feature of the Hamiltonian. For $q = 0.2$ nm$^{-1}$ (red dashed line), the Drude peak is shifted to higher energy and separated into two peaks in the frequency region $\omega < 0.2E_0$. For $q = 0.4$ nm$^{-1}$ (green dash-dotted line), only one peak is in the frequency region $\omega < 0.2E_0$, and the other peak shifts to the interband region $\omega > 0.2E_0$. The interband peaks should be compared with the peaks in the joint DOSs for $\text{Re}(q = 0, \omega)$ or the joint spectral function for $\text{Re}(q \neq 0, \omega)$.

the same $q$, we observe two intraband peaks roughly at $\omega_1 = q \times v_F$ and $\omega_2 = q \times v_F$ in the frequency region $\omega < 0.2E_0$. The interband region starts from approximately $\omega > 0.2E_0$, as observed in the optical conductivity for the case $q = 0$. The peaks in the interband region are determined from the peaks in the joint DOSs. For the case $q \neq 0$, the peaks in the interband region are determined from the peaks in the joint spectral functions.

### A3 Band structure and a tight binding model of the monolayer VSe$_2$

In this section, we present the DFT calculations of the 2D and 3D VSe$_2$ band structures and compare them with the results presented in the main text. In Figure a2, we present the DFT calculation with the SOC effect and observe that the impact of the SOC on 1T-VSe$_2$ is minimal. In Figure a3, we present the 3D VSe$_2$ band structure. A simplified tight binding model could be obtained using the hopping parameters in Tables 1-3 of the main text. In Figure a4, we use a schematic to describe the hoppings on the lattice. The solid line represents the hopping between the V and Se atoms. The hopping parameters are given in Table 2 of the main text. The model is given by:

$$H_t = \sum_{i,d,p} t_{dp1}[V_{id} \langle S_{e_{1,\delta,p}} \rangle + \langle S_{e_{1,\delta,p}} \rangle V_{id}] + t_{dp2}[V_{id} \langle S_{e_{2,\delta,p}} \rangle + \langle S_{e_{2,\delta,p}} \rangle V_{id}]$$

where $i$ is the lattice site, $\delta = \delta_{1,2,3}$ are the three hopping vectors, $d$ represents the five $d$ orbitals of the V atom, $p$ represents the three $p$ orbitals of the Se atom, and $t_{dp1}$ and $t_{dp2}$ are the hopping parameters given in Table 2 of the main text. Here, $\delta_1 = (1.667, 0.962), \delta_2 = (0.000, -1.924)$, and $\delta_3 = (-1.667, 0.962)$ in the unit of Å. The tight binding

![Figure a2](image2.png)

**Figure a2** Band structure of the monolayer VSe$_2$ from the DFT calculation with spin-orbit coupling (SOC). The band structure is close to the spin-polarized DFT calculation, as shown in Figure 2(a) of the main text. In the 2D calculation, we set the lattice constant $c = 20$ Å to ensure that there is no interaction between the layers to achieve an ideal single-layer 2D material condition.

![Figure a3](image3.png)

**Figure a3** (Color online) Band structure of 3D VSe$_2$ from the spin-polarized DFT calculation. The band structure around the points Γ, M, K is close to those in the 2D monolayer VSe$_2$ (Figure 2(a) of the main text). In 3D, we have more high symmetry momentum points H, A, and L and observe different band structures around these points as expected.

![Figure a4](image4.png)

**Figure a4** (Color online) Simplified tight binding model based on the Wannier fitting of the band structure of the monolayer VSe$_2$. The solid line represents the hopping between the V and Se atoms, and the hopping parameters are given in Table 2 of the main text. The dashed line represents the hopping between the Se$_1$ and Se$_2$ atoms. The hopping parameters are given in Table 1 of the main text. Note that the hopping terms in the dashed lines are not included in the approximate tight binding model $H_t$. 

![Figure a5](image5.png)
Hamiltonian is an $11 \times 11$ matrix. The dashed line in Figure a4 represents the hopping between the Se$_1$ and Se$_2$ atoms. The hopping parameters are given in Table 1 of the main text. The next-nearest neighbor hopping between the Se atoms could be obtained from Table 3 of the main text. In Table a1 of this Appendix, we list the nearest-neighbor V-V hopping parameters for VSe$_2$ from the cell (000) to (100), which is much smaller than the other hopping parameters.

| V(1 0 0)          | $d_{xy}$ | $d_{xz}$ | $d_{yz}$ | $d_{xz}$ | $d_{y^2}$ |
|------------------|----------|----------|----------|----------|-----------|
| $V_{11}$         | -0.183   | 0        | -0.052   | 0.153    | 0         |
| $V_{22}$         | 0        | -0.052   | 0        | 0        | 0.215     |
| $V_{33}$         | -0.052   | 0        | 0.095    | -0.085   | 0         |
| $V_{44}$         | 0.153    | 0        | -0.085   | -0.271   | 0         |
| $V_{55}$         | 0        | 0.215    | 0        | 0        | 0.135     |