Topologically nontrivial interband plasmons in type-II Weyl semimetal MoTe₂

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Abstract

In many realistic topological materials, more than one kind of fermions contribute to the electronic bands crossing the Fermi level, leading to various novel phenomena. Here, using momentum-resolved inelastic electron scattering, we investigate the plasmons and their evolution across the phase transition in a type-II Weyl semimetal MoTe₂, in which both Weyl fermions and trivial electrons contribute to the Fermi surface in the $T_d$ phase. One plasmon mode in the $1T'$ phase at high temperature and two plasmon modes in the topological $T_d$ phase at low temperature are observed. Combining with first-principles calculations, we show that all the plasmon modes are dominated by the interband correlations between the inverted bands of MoTe₂. Especially in the $T_d$ phase, since the electronic bands split due to inversion symmetry breaking and spin–orbit coupling, the plasmon modes manifest the interband correlation between the topological Weyl fermions and the trivial electrons. Our work emphasizes the significance of the interplay between different kinds of carriers in plasmons of topological materials.

1. Introduction

Topological materials including topological insulators [1–4] and topological semimetals [1–5] have become an extremely attractive branch in condensed matter physics. The nontrivial topology of Bloch bands could lead to a variety of novel physical phenomena such as the topologically protected edge states and the related unique transport and optical behaviors [6], leading to potential technological applications in spintronics and quantum computations. The collective excitation of electrons due to their long-range Coulomb interaction, i.e., plasmon, is a fundamental concept in physics, and has attracted increasing attentions in the field of topological materials recently. Various novel plasmons have been predicted in topological insulators [7–10] and in topological semimetals [11–25]. For example, the surface states of three-dimensional topological insulators with the spin–momentum locking was predicted to support the spin plasmon, linking spintronics and plasmonics [7]. The Weyl semimetals without time-reversal symmetry were predicted to host the nonreciprocal magnetoplasmon, which is the signature of the chiral anomaly [15]. Importantly, a series of significant experimental progresses on plasmon excitations have been made in topological...
insulators [26–30] as well as in topological semimetals [31–34], which could greatly facilitate the promising applications of these topological materials in plasmonics [35].

Most of these fascinating plasmons in topological materials can be well described within a single band picture with only one kind of carriers involved. This is similar to the cases in normal metals, in which the intraband correlation dominates [36]. Consequently, the origin of these plasmons can be directly understood in terms of the simple Fermi surfaces. However, in many topological materials, such as NbP and (W, Mo)Te₂, the topologically nontrivial bands and the trivial bands coexist around the Fermi level, and thus more than one kind of fermions contribute to the Fermi surface [37–46]. As the result, the plasmon modes in these systems may be contributed from multiple kinds of fermions. Especially, when the topologically nontrivial and trivial bands highly mix with each other, their couplings could significantly affect the properties of the plasmons, which is beyond the scope of the single band picture. Therefore, the exotic plasmon properties based on the assumption of pure topological bands may not be applicable in real topological materials.

In this letter, using high-resolution electron energy loss spectroscopy (HREELS) [47], we studied the plasmons in MoTe₂, which exhibits the structural phase transition from the high temperature monoclinic $1T'$ phase to the orthorhombic $T_d$ phase with the critical temperature of $\sim 250$ K. We observe one plasmon mode in the $1T'$ phase and two plasmon modes in the $T_d$ phase. The energies of these plasmon modes are almost dispersionless and all exhibit strong temperature dependences. Combining with first-principles calculations, we reveal that unlike the conventional plasmons dominated by the intraband correlations, all these plasmon modes in MoTe₂ mainly originate from the interband correlations. Especially in the $T_d$ phase, the two plasmon modes are dominated by the correlations between the nontrivial Weyl fermion bands and the trivial electronic bands. Our work clearly demonstrates that the interplay of topologically trivial electrons and Weyl fermions plays a crucial role in plasmons of realistic topological materials.

2. Materials and methods

2.1. Crystal preparation

Single crystal of $1T'$-MoTe₂ were grown by using Te as flux. Starting materials Mo (column, 99.9999%) and Te (lump, 99.9999%) were mixed in an Ar-filled glove box at a molar ratio of Mo:Te = 1:20. The mixture was placed in an alumina crucible, which was then sealed in an evacuated quartz tube. The tube was heated to 1100 °C over 20 h and dwelt for 10 h. Then, the tube was slowly cooled down to 950 °C at a rate of 1 °C h⁻¹ followed by separating the crystals from the Te flux by centrifuging. Shiny crystals with the size of $1 \times 5 \text{ mm}^2$ were obtained on the bottom of the crucible.

The good crystal quality is characterized ex situ by x-ray diffraction and the surface of the cleaved sample is checked by x-ray photoelectron spectroscopy (XPS), which can exclude the possible surface contamination or oxidation of them. The details of the sample characterizations are shown in the supplemental materials (https://stacks.iop.org/NJP/22/103032/mmedia).

2.2. HREELS measurement

As a surface sensitive technique, HREELS is an ideal candidate to explore the low-energy collective excitations of MoTe₂. Compared with conventional HREELS, our recently developed two-dimensional (2D)-HREELS can directly obtain a 2D energy-momentum mapping in a very large momentum scale without rotating sample, monochromator, or analyzer [48].

The energy and momentum of the collective excitations (either plasmon or phonon) are obtained using the conservation of energy and momentum for incident and scattered electrons. As given by $h\mathbf{q}_i = mE_i \sin \alpha_i = k_i \sin \alpha_i$ (where $\alpha_i$ and $\alpha_s$ are the incident and scattering angles, respectively), the parallel momentum $q_i$ depends on incident energy $E_i$, energy loss $E_{\text{loss}} = E_s - E_i$, $\alpha_i$ and $\alpha_s$ according to

$$q_i = \frac{\sqrt{2mE_i}}{h} \left( \sin \alpha_i - \sqrt{1 - \frac{E_{\text{loss}}}{E_i}} \sin \alpha_s \right) \approx \frac{\sqrt{2mE_i}}{h} (\sin \alpha_i - \sin \alpha_s). \quad (1)$$

In this study, all the HREELS measurements were performed in situ within $\sim 10$ h after fresh cleavage in ultra-high vacuum ($\sim 1 \times 10^{-10}$ Torr). We obtained the information around the first Brillouin zone (BZ) center $\Gamma$ with the incident energy ranging from 15 eV to 110 eV at room temperature. And the temperature-dependent measurements were obtained with the incident energy of 110 eV only.

2.3. Details of first-principles calculations

The first-principles calculations are performed by using Vienna $ab$ initio simulation package (VASP) [49] based on the density function theory with Perdew–Burke–Ernzerhof parameterization of generalized
The effective background dielectric constant $\kappa$ is calculated from VASP, the values of which are 218.2 for the $T_d$ phase and 190.3 for the $1T'$ phase along the $\Gamma-X$ direction ($\kappa_{\omega}$). The formula of $\varepsilon^{\mathrm{RPA}}(q, \omega)$ can be written as the long wavelength limit of $q = 0$:

$$\varepsilon(0, \omega) = 1 + \frac{4\pi\varepsilon^2}{\kappa N_k} \left[ \sum_{m,k} \left( \frac{\partial E_{mk}}{\partial W_{mk}} \right)^2 \right] - \sum_{m,k \neq m} \frac{f_{mk} - f_{nk}}{E_{mk} - E_{nk} + i\eta_2} \left( \langle mk | \frac{\partial E}{\partial W} | nk \rangle \langle nk | \frac{\partial E}{\partial W} | mk \rangle \right)$$

where the first term is the intraband term with a broadening parameter $\eta_1$, and the second term is the interband term with a broadening parameter $\eta_2$.

3. Results and discussions

3.1. Crystal structures of MoTe$_2$

MoTe$_2$ is a layered Van der Waals material with two different structures at 300 K: hexagonal ($2H$) phase or monoclinic ($1T'$) phase, due to different growth conditions. In this study we focus on the $1T'$ phase (a trivial metal/semimetal), which exhibits a structural phase transition to the $T_d$ phase at $\sim 250$ K [53]. Although these two phases share the same in-plane structure, the $T_d$ phase with a vertical (90$^\circ$) stacking belongs to the non-centrosymmetric space group $Pmn2_1$, while the $1T'$ phase with a distorted stacking belongs to the centrosymmetric space group $P21/m$ [see figures 1(a) and (c)]. Only the low temperature $T_d$ phase was predicted to support type-II Weyl fermions, while the centrosymmetric $1T'$ phase is topologically trivial. Angle-resolved photoemission spectroscopy together with the first-principles calculations shows that Fermi surfaces in the $T_d$ phase MoTe$_2$ consist of both topologically trivial and nontrivial bands [41–44, 46]. Figure 1(e) shows the BZ of the $T_d$ phase, and the locations of the Weyl points are shown in figure 1(f) and a corresponding enlarged zone in figure 1(g). Previous theoretical and experimental works suggest four pairs of Weyl points in the $k_z = 0$ plane of the BZ [37, 38].

The MoTe$_2$ single crystals with the $1T'$ phase at $\sim 294$ K were cleaved in situ, and the obtained (001) surface exhibits excellent (1 $\times$ 1) low energy electron diffraction (LEED) patterns of sharp spots with a very low background, as shown in figure 1(b). At 44 K, the $T_d$ phase surface shows almost unchanged (1 $\times$ 1) LEED patterns [figure 1(d)] as the $1T'$ phase, since the two phases share the same in-plane structure. The angle-resolved inelastic electron scattering was performed by the HREELS with the capability of 2D energy-momentum mapping [48]. The results in both the $1T'$ and $T_d$ phases at different temperatures were obtained.

3.2. Plasmons from HREELS measurements

Figures 2(a) and (b) display the 2D energy-momentum mappings obtained from the HREELS measurements along the $\Gamma-X$ direction with the incident beam energy of 110 eV for the $T_d$ phase at 44 K and for the $1T'$ phase at 294 K, respectively. In the $T_d$ phase, the energy distribution curve (EDC) integrated over momentum [red curve in figure 2(a)] shows two distinct energy-loss peaks around 90 and 170 meV, which are labeled as $\alpha$ and $\beta$, respectively. In contrast, the integrated EDC for the $1T'$ phase only shows one energy-loss peak around 220 meV, which is labeled as $\gamma$. The energies of all these peaks are much higher...
than the highest phonon energy (around 35 meV) in MoTe$_2$ [54], indicating that they are not phonons. Through XPS measurement to test the 3d binding energy of Te and Mo, and the comparison of the HREELS spectra between clean and exposed surfaces, we show that these loss peaks are not vibrations of possibly adsorbed molecules (e.g., H$_2$O) or the Te–O bond due to possible surface oxidation. Instead, they are most likely to be plasmons.

Then the dispersions of these modes are checked from $q$-dependent EDCs extracted from the 2D mapping, as shown in figures 2(c) and (d). Due to the semimetallic nature of MoTe$_2$, there exists a strong Drude background in the energy loss EDCs. We employ a background subtraction method based on a polynomial fitting of the baseline, which has been used in graphene [55] and graphite [56], to extract the information of the exact energy loss peaks, including energy, full width at half maximum, and intensity (see details in the supplemental materials). The exact fitted energies of $\alpha$, $\beta$ and $\gamma$ modes at the $\Gamma$ point are $89(\pm1)$ meV, $174(\pm6/-3)$ meV and $216(\pm13/-19)$ meV, respectively. The obtained experimental energy-momentum points of these modes are plotted in figures 2(e)–(h) (solid dots). All of the three modes are almost dispersionless, and their intensities show quick damping with the increasing momentum $q$, becoming invisible when $q > 0.05$ Å$^{-1}$. In addition, their energies are nonzero at $q = 0$, evidencing that they are not plasmons originating from surface states. Instead, they should be bulk (bp) or surface plasmons (sps) originating from the bulk bands. In the $T_d$ phase, the average ratio of energy between $\alpha$ and $\beta$ is $\frac{\omega_{\alpha}}{\omega_{\beta}} \sim 0.56 \pm 0.04$, significantly deviating from the conventional ratio between the energies of bp and its corresponding sp from the same electronic band: $\frac{\omega_{\alpha}}{\omega_{bp}} = \frac{1\sqrt{2}}{2} \sim 0.71$ for the ideal case, and varies from 0.60 $\pm$ 0.35 (lithium) to 0.73 $\pm$ 0.11 (potassium) for typical real metals [57]. And in the 1$T'$ phase, only one mode ($\gamma$) is detected. These observations suggest that $\beta$ and $\alpha$ cannot be the conventional bp and its corresponding sp. However, our current results with different incident angles and various incident energies (shown in the supplemental materials) cannot provide convinced evidence if the observed loss features are bp or sps.

To further clarify the nature of these modes, we performed first-principles calculations to obtain the electronic bands as well as the density–density correlation functions in the two phases of MoTe$_2$. Then the dynamical dielectric functions $\varepsilon_{\text{RPA}}(q, \omega)$ were calculated within the RPA. The calculated plasmon modes appear as sharp peaks in the energy loss function $\varepsilon_{\text{loss}} = -\text{Im}(\varepsilon_{\text{RPA}})$, which are directly related to the experimental excitation spectra probed by HREELS. First, we checked the bulk or surface nature of the modes by performing a slab calculation (details in the supplemental materials). It turns out that the peak positions of the energy loss function in the slab center (corresponding to bulk modes) are almost identical to those obtained at the slab surface (corresponding to surface modes). As a result, no matter if the measured plasmon modes are surface or bulk, their originations are the same, both from the bulk band structure.
In addition to the terms describing the intraband transitions in the regular plasmon study, the calculations also include the terms describing the interband transitions which may dominate in the plasmons with inverted energy bands [9]. We set the Fermi energy at $-50$ meV, and use two different broadening parameters for the intraband ($\eta_1 = 150$ meV) term and the interband ($\eta_2 = 20$ meV) term, respectively, to fit the experimental results. It turns out that the interband transitions dominate the loss function intensities, while the intraband transitions only act as negligible backgrounds. We will focus on the interband contributions in the following. The intensity mappings of the calculated electron energy loss functions for the $T_d$ phase and the $1T'$ phase are shown in figures 2(e) and (f), and the corresponding loss functions at different $q$ are plotted in figures 2(g) and (h), respectively. The experimental energy-momentum points of the three modes are superimposed on the calculated results. Considering the experimental energy uncertainty shown above, the theoretical simulation in the small $q$ range shows a good agreement with the measured peak positions. The main discrepancy is that the calculated results damp slower and disperse to a larger momentum range where the loss peaks are invisible in the measurements. This is largely due to the simplification of the theoretical model. In the simulations, all the plasmon modes are simplified as interband correlations, with the intraband correlations neglected, which might also contribute to the damping rate. Other scattering processes, such as electron–phonon scattering, electron-defect scattering and electron-surface scattering, are also ignored in the simulations, resulting in the discrepancy of the momentum of electron–hole excitations between the simulation and experimental observations. Nevertheless, the theoretical calculations can well reproduce the energies of the observed plasmon modes as well as their connections to the phase transition. Details of the calculation methods and analyses can be found in the supplemental materials.

3.3. Temperature dependence of plasmons

To gain more insights into the behaviors of plasmons accompanied with the structural phase transition, we performed temperature-dependent HREELS measurements from 294 K to 44 K. Figure 3(a) shows the stacked EDCs at $\Gamma$ point at several temperatures (a complete set of temperature-dependent measurements is
provided in the supplemental materials]. With the temperature decreasing, the γ mode gradually evolves into the β mode, while the α mode gradually appears. The energies and intensities of these three modes as a function of temperature are extracted by using the fitting method mentioned above, with the results plotted in figures 3(b) and (c). When the temperature is above ~260 K, the data can be only well fitted with one peak. And when the temperature is below ~200 K, the data can be only well fitted with two peaks. It should be noted that, in the temperature range from 200 K to 260 K where the structural phase transition temperature $T_C$ locates, the data can be fitted equally well either with one peak or with two peaks. The present HREELS plasmon measurements cannot provide accurate information near $T_C$, possibly because of the intermediate new phase with frustrated disorders [58] in the structure associated with the phase transition. But it is pretty clear that only the γ mode exists in the high-temperature $1T'$ phase, while there are indeed two modes (α and β) existing in the low-temperature $1T_d$ phase.

Figure 3(b) shows that the energies of the observed plasmon modes exhibit similar temperature-dependent behaviors, all increasing with rising temperature. This is different from the prediction that the energy of the intrinsic plasmon in type-I Weyl/Dirac semimetals has a nonlinear temperature dependence when only a single Weyl/Dirac cone contributes to the Fermi surface [13, 14]. The electronic structure of MoTe$_2$ is more complicated than the single Weyl/Dirac cone model, and all of the three plasmon modes mainly originate from the interband correlations. As a result, the temperature dependences of energies of these modes should be hard to be faithfully captured by the theoretical predictions based on the simple ideal model.

3.4. Origin of the interband plasmons
To explore the origin of these plasmon modes, we decompose the contribution of each plasmon mode to the energy loss function [i.e., $\text{Im} \varepsilon^{\text{RPA}}(0, \omega)$] (with details described in the supplemental materials) and project it onto the energy bands along a selected path in the BZ. Figures 4(a)–(c) show the distributions of $\text{Im} \varepsilon^{\text{RPA}}(0, \omega)$ for α, β and γ in the BZ, respectively. The distributions of $\text{Im} \varepsilon^{\text{RPA}}(0, \omega)$ for the α and β modes are different, and those for the β and γ modes share some similarity, while they are still distinct due to the difference of the band structures in the two phases. Figures 4(d)–(g) illustrate the band structures, contributions and the corresponding strength of $\text{Im} \varepsilon^{\text{RPA}}(0, \omega)$ for the three modes in the $1T_d$ phase and $1T'$ phase along the $\Gamma$–$P$ direction and $Q$–$Q$ direction, respectively. The relevant bands are labeled as $a$, $b$, $c$ in the $1T'$ phase and $a_1$, $a_2$, $b_1$, $b_2$, $c_1$, $c_2$ in the $1T_d$ phase, respectively. As shown in figure 4(f), the band $a$ and the band $c$ can be regarded as two crossing bands with a band gap under the influence of the spin–orbit coupling. In this kind of inverted band structure, a plasmon mode γ originating from the interband transitions indeed exists, similar to the case in reference [9]. Accompanied with the structural phase transition from the $1T'$ phase to the $1T_d$ phase, the bands $a$, $b$, $c$ split into $a_1$/$a_2$, $b_1$/$b_2$, and $c_1$/$c_2$ due to the inversion symmetry breaking, forming eight Weyl points at the crossing points of the bands $a_1$/$a_2$ and $b_1$/$b_2$. The positions of the Weyl points W1 and W2 are shown in figure 4(e). Accordingly, the γ mode in the $1T'$ phase splits into two plasmon modes, α and β. The interband correlations between $a_2$ and $c_2$ contribute to the α mode while the correlations between $a_1$ and $c_1$ contribute to the β mode [shown in figure 4(d)].
These two plasmon modes in the $T_d$ phase are dominated by the interband correlations between the topologically nontrivial bands and trivial bands.

4. Conclusions

In conclusion, we have systematically investigated the plasmon modes of MoTe$_2$ at different temperatures. Our HREELS experiments indicate the presence of two plasmons ($\alpha$ and $\beta$) in the low temperature $T_d$ phase and only one mode ($\gamma$) in the high temperature $1T'$ phase. Combining with first-principles calculations, we find that all the modes are dominated by interband correlations between the inverted bands of MoTe$_2$. Especially, the modes in the topological $T_d$ phase, dominated by the interband correlations between the topologically nontrivial bands and trivial bands, are attributed to the band splitting due to inversion symmetry breaking. This work reveals the limitation of the single band picture and significantly broadens the understanding of plasmon modes in realistic topological materials, in which band mixing usually exists.

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