Temperature induced band shift in ferromagnetic Weyl semimetal Co$_3$Sn$_2$S$_2$

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The discovery of nonmagnetic Weyl semimetals (WSMs) in TaAs compounds has triggered lots of efforts in finding its magnetic counterpart. While the direct observation of the Weyl nodes and Fermi arcs is a magnetic candidate through angle-resolved photoemission spectroscopy is hindered by the complex magnetic domains. The transport features of magnetic WSMs, including negative magnetoresistivity and anomalous Hall conductivity, are not conclusive since these are sensitive to extrinsic factors like defects and disorders in lattice or magnetic ordering. Here, we systematically study the temperature dependent optical spectra of ferromagnetic Co$_3$Sn$_2$S$_2$ experimentally and simulated by first-principles calculations. The many-body correlation effect due to Co 3d electrons leads to renormalization of bands by a factor about 1.33, which is moderate and the description within density functional theory is suitable. As temperature drops down, the magnetic phase transition happens and the magnetization drives the band shift through exchange splitting. The optical spectra can well detect these changes, including the transitions sensitive and insensitive to the magnetization, and those from the bands around the Weyl nodes. The results strongly support that Co$_3$Sn$_2$S$_2$ is a magnetic WSM and the Weyl nodes can be tuned by magnetization with temperature change.

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Recently, the shandite compound Co$_3$Sn$_2$S$_2$ has attracted lots of attentions since it not only shows intrinsic ferromagnetism but also is proposed to have three pairs of Weyl points around the Fermi level in the first Brillouin zone (BZ) [1–3]. For its quasi-two-dimensional crystal structure, low carrier density and strong anomalous Hall effect (AHE), Co$_3$Sn$_2$S$_2$ has been thought as a potential candidate to realize the quantum AHE [4–7] in its tow-dimensional (2D) limit [8,9]. As a candidate of magnetic Weyl semimetal (WSM) [10], the magnetic ordering states are expected to have interactions with Weyl nodes [2,3], so that the topological properties from WSM can be finely tuned through control of magnetization [2,11]. Actually, it has been found that upon cooling, the magnetization and anomalous Hall conductivity (AHC) of Co$_3$Sn$_2$S$_2$ vary accordingly [1,2] and the first-principles calculations of AHC at different magnetization has shown their intrinsic relationship through the changes in the position of Weyl nodes [2]. To check whether this picture is true or not in realistic samples, here we have performed systematical optical spectra measurements under different temperatures ($T$s). The advantages of optical spectra measurements over the AHC measurements are as following: First, optical spectrum is sensitive to both occupied and unoccupied bands, which constitute the Weyl nodes in WSM. In contrast, AHC is only sensitive to the one crossing Fermi level [7,13–17]. Second, on considering optical selection rule and peak position, one can definitely identify the contributions from Weyl nodes [18,20]. Third, transport measurements like AHC requires high techniques and high quality of the whole device [15,17]. The extrinsic factors make the understanding of intrinsic mechanism very difficult.

In this work, combining optical spectroscopy and first-principle calculations, we systematically investigate the band structure of Co$_3$Sn$_2$S$_2$ at various $T$s above and below its Curie temperature ($T_C$). We have clearly demonstrated how the band structure evolves and identified both the magnetization sensitive and nonsensitive contributions to different optical conductivity peaks. The contributions around the Weyl nodes are also identified. Furthermore, we have found that the electron-electron correlation effect in this magnetic compound is not strong and the band renormalization factor is around 1.33. The first-principles calculations within local density functional approximation is suitable. These give out strong supports that Co$_3$Sn$_2$S$_2$ is a magnetic WSM.

The $T$ dependence of the real part of the in-plane optical conductivity [$\sigma_1(\omega)$] is shown in the energy range from $20$ cm$^{-1}$ (2.5 meV) to $7000$ cm$^{-1}$ (0.87 eV) (Fig. 1, see details of the measurements in the supplementary material [21]). The conductivities at 100 K (blue) and 5 K (black) are shown in the infrared region for comparison.

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At first glance, they consist of a Drude peak at zero-frequency and several other Lorentz peaks at finite frequencies. Besides, several sharp features in conductivities are observed at 155 cm^{-1}, 245 cm^{-1} and 370 cm^{-1}, which comes from the infrared-active vibrations. Based on Kubo function, σ₁(ω) is proportional to the joint density of states, the peak at zero frequency (Drude) represents the intra-band response, and the one at a finite frequency (Lorentz) comes from the inter-band transition \([12]\). The extrapolated values for the dc conductivity \(\sigma_{\text{dc}}(0) = \sigma_{\text{dc}}(\omega \rightarrow 0)\) are virtually identical to the resistivity, indicating excellent agreement between optical and transport measurements. Upon cooling, \(\sigma_1(\omega)\) shows apparent \(T\) dependence, especially in the mid-infrared range, indicating that the band structure varies prominently with decreasing \(T\).

At room temperature (300 K), the optical conductivity shows a typical metallic character, with a Drude-like free-carrier response at zero frequency. Besides a Lorentz-like feature centered around 5 000 cm^{-1} and some phonon peaks, in the range from 1 000 to 3 000 cm^{-1} the optical conductivity is almost frequency independent. This plateau persists down to \(T_C \sim 177 \text{ K}\), low which it gradually evolves into a Lorentzian, which grows up and moves to lower energy range with decreasing \(T\). Meanwhile, the peak centered at 5 000 cm^{-1} starts to strengthen and moves to higher energy. These features indicate that, in the FM state, the band structure changes dramatically with \(T\). The Drude peak at zero frequency narrows continuously with decreasing \(T\), implying suppressed quasiparticle scattering. Below 100 K, the intra-band response is greatly suppressed, evolving into two components: an ultra-sharp Drude peak and a new interband absorption peak at 310 cm^{-1} (inset of Fig. 1c). In the corresponding reflectivity (Fig. S1 in the supplementary material [21]), a sharp plasma edge emerges below 100 K, indicating much coherent quasiparticle response [22, 23].

From the spectral weight of \(\sigma_1(\omega)\) (Fig. 1d), we see that, from 300 to 200 K, the \(S(T, \omega)\) is gradually transferred from high to low energy range, indicating an enhanced intra-band response [21]. While, below \(T_C \sim 177 \text{ K}\), spectral weight below 1000 cm^{-1} is transferred back to high energy range, accumulating in peaks at 2000 and 5000 cm^{-1}, respectively. Correspondingly, in Fig. 1c, upon cooling, the spectral weight at the far-infrared range is gradually transferred to the mid-infrared absorption peaks (i.e., interB and interC). However, below 100 K, the spectral weight of low-energy intra-band response is greatly suppressed, giving rise to a new absorption peak around 310 cm^{-1} (38 meV, interA in the inset of Fig. 1c). Although such behavior is similar to the spin/charge-density-wave transition [25, 26], during which a gap emerges on the Fermi surface, in \(\text{Co}_3\text{Sn}_2\text{S}_2\), no phase transition was predicted and observed in previous studies [11, 12].

To get some more quantitative estimates of parameters determining the optical response, we fit the optical conductivity using a Drude-Lorentz model with the dielectric function \(\varepsilon = \varepsilon_{\infty} + i \varepsilon_2\) [12, 27]

\[
\varepsilon(\omega) = \varepsilon_{\infty} - \sum_i \frac{\omega^2_{p,D,i}}{\omega^2 + \frac{i\omega}{\tau_{D,i}}} + \sum_j \frac{\Omega_j^2}{\omega^2_j - \omega^2 - i\omega\gamma_k},
\]

in which \(\varepsilon_{\infty}\) is the real part at high frequency. In the first sum \(\omega^2_{p,D,i} = 4\pi n_i e^2/m_i^*\) and \(1/\tau_{D,i}\) are the square of plasma frequency and scattering rate for the delocalized (Drude) carriers, respectively, and \(n_i\) and \(m_i^*\) are the carrier concentration and effective mass. In the second summation, \(\omega_j, \gamma_k\) and \(\Omega_k\) are the position, width and strength of the \(j\)th vibration or bound excitation. The complex conductivity is \(\sigma(\omega) = \sigma_1 + i\sigma_2 = -2\pi i\omega(\varepsilon(\omega) - \varepsilon_{\infty})/Z_0\) (\(Z_0 \approx 377 \Omega\) is the impedance of free space), and the real part \(\sigma_1(\omega)\) is fitted using a non-linear least-squares technique.

The fit to the data at 100 K shown in Fig. 2b indicates that the optical conductivity can be reproduced quite well using one Drude peak and three Lorentz oscillators at 310, 1600 and 5100 cm^{-1} (\(\approx 38, 198\) and 632 meV). Below 100 K, Drude response is suppressed dramatically, revealing the absorption peak centered at 310 cm^{-1} (Inset of Fig. 1c). We realize that it’s hard to describe this
Here, we notice that even though previous investigations on Co$_3$Sn$_2$S$_2$ predict the emergence of Weyl points right below $T_C \sim 177$ K [18, 20, 28, 29](see more discussion in Supplementary material [21]). When SOC is further considered, the crossing points open gaps along the nodal rings in the mirror planes [2]. On considering SOC, band gap opens along the rings. Fig. 3(b) and (c) are band structures in the cases of 0.15 $\mu_B$ and 0.33 $\mu_B$ on Co atoms, respectively. The latter one is in the ground state without any constraint. The spin exchange splitting gradually separates the bands into spin up and spin down channels. The spin-up one shifts closer to the Fermi level, and the spin-down moves to higher energy. In the case without SOC (Fig. S5), the nodal rings protected by mirror plane are kept well in both channels. The Fermi surfaces in PM and FM are different as shown in Figs. S2 (d) and (e). It becomes a half-metal in the ground state (Fig. S5b in the supplementary material [21]). When SOC is further considered, the crossing points open gaps along the nodal rings and away from the mirror planes, three pairs of Weyl points appears. The band structure around Weyl point is described in Fig. S2 [21] and Ref. [2].

Next, we calculated the optical conductivity with different magnetization. In addition to the inter-band contributions, the intra-band transition is included as the Drude model. The plasma frequency $\omega_p$ contributed to the Drude peak can be obtained from band structure calculations. It is obtained as 1.73 eV for PM and 1.34 and 0.94 eV for FM with 0.15 and 0.33 $\mu_B$/Co, respectively. $\omega_p$ decreases with magnetization, which is consistent with the shrinking of Fermi surfaces in Figs 3(d) and (e), as well as the decreasing of carrier density [1]. The scattering rate $\frac{1}{\tau_D}$ is parameterized as 0.17 eV, 0.05 eV and 0.03 eV to mimic the decreasing (increasing) of $T$ (lifetime) and to well fit the experimental spectra. Within these settings, the theoretical spectra are shown in Fig. 3(f) together with the experimental ones. It is noted that the experimental spectra are replotted with the incident photon wavenumber being scaled by a factor of 1.33 to match the InterC in Fig. 3 with the calculated position.

To better understand the optical conductivity as well as the electronic properties, we have simulated the band structure and optical conductivity of Co$_3$Sn$_2$S$_2$ through the first-principle calculations. Because of the weaker magnetic fluctuation as $T$ drops, the effective moment on Co atoms increases upon cooling. To mimic the $T$ effect, the magnetic moment on Co is constrained at different values during the self-consistent calculation. In paramagnetic (PM) state, the calculated band structure shows semi-metallic feature in Fig. 3a). Without including SOC, there exist two linear band crossings marked in dashed circles in $U-L$ and $L-T$ paths as protected by the mirror symmetry. These crossing points are on the nodal rings in the mirror planes [2].

Figure 2. (a)The Drude-Lorentz fit to the complex optical conductivity of Co$_3$Sn$_2$S$_2$ at 100 K. (b)The optical conductivity at 5 K, after the Drude peak and phonon peaks below 300 cm$^{-1}$ have been subtracted. Inset: the whole optical conductivity at 5 K, the green dashed line represents the intra-band response (Drude), origin line is the fit to the phonon modes below 300 cm$^{-1}$ (with a series of Lorentz peaks). (c) and (d) are the fitted value for the square of plasma frequency($\omega_{p,D}^2$) and the scattering rate($\gamma$) of Drude component, the red line in (c) is the $T^2$ fit to the square of plasma frequency. The dashed lines in (c) and (d) denotes the Curie Temperature ($T_C \sim 177$ K) and 100 K.

peak with a simple Lorentz peak for its steep edge on the low-energy side. To investigate this low-energy interband transition, in Fig. 2b, we have subtracted the Drude response and sharp phonon peaks (inset of Fig. 2b) [28]. In the residual conductivity, which purely comes from interband transitions, an unusual feature is a $\omega$-linear conductivity from 130 to 230 cm$^{-1}$ (16-29 meV), which indicates the presence of 3D linear bands near the Fermi level [18 20 28 29](see more discussion in Supplementary material [21]).

The $T$ dependence of the square of plasma frequency ($\omega_{p,D}^2$), i.e., the Drude weight, and scattering rate ($1/\tau_D$) of Drude response are shown in Figs. 2c and d, respectively. As the temperature is reduced, the Drude weight is firstly enhanced a little at 200 K and then suppressed after the FM transition. Below 100 K, $\omega_{p,D}^2$ is considerably suppressed and shows $T^2$ dependence, which agrees well with theoretical predictions for Weyl semimetals [30]. The $1/\tau_D$ decreases dramatically with the $T$, from 495 cm$^{-1}$ at 300 K to 15 cm$^{-1}$ at 5 K. Here, we notice that even though previous investigations on Co$_3$Sn$_2$S$_2$ predict the emergence of Weyl points right below $T_C \sim 177$ K [18], we only observed the distinct signature of linear bands at $T < 100$ K. The optical response indicates that, above 100 K, the response from other parabolic bands may superimpose over the response from the bands with linear dispersion. Below 100 K, the parabolic bands may gradually move away from Fermi level, resulting in a Lifshitz transition, which leaves the linear bands dominating the low-energy response. At 5 K, both the steep plasma edge in reflectivity (Fig. S1 in supplementary material [21]) and the ultra-narrow Drude peak in optical conductivity reflect the coherence of the linear bands [18 19].
Figure 3. (a) The band structure along high-symmetry paths with (red) and without (black) spin-orbital coupling (SOC). The dashed circles denote the band crossing opened by SOC. (b) and (c) show the band [0.15\(\mu_B\)/Co (b), 0.33\(\mu_B\)/Co (c)]. Comparing with the measured optical conductivity, we ascribe the three absorption peaks in Fig. 1c to inter-band transitions 'interA', 'InterB' and 'InterC', respectively. The band structures in (b) and (c) are calculated with SOC. (d) and (e) are Fermi surfaces in paramagnetic (PM) and ferromagnetic (FM, 0.33\(\mu_B\)/Co) state. (f) displays the simulated optical conductivities (solid lines) based on band structure calculation (with SOC) in the PM and FM (0.15\(\mu_B\)/Co and 0.33\(\mu_B\)/Co) states in comparison with experimental results (dashed dot lines). The energy discrepancy between the experimental and theoretical results mainly comes from the correlation effect, which is not considered during the calculation.

This difference between the experimental and theoretical results comes from the correlation effect due to 3d orbitals of Co, which is not considered during the calculation within local density approximation. The overall consistency between the calculated and scaled experimental spectra indicates that the single-particle approximation can catch the most essential features of this material and the electron correlation is moderate in Co\(_3\)Sn\(_2\)S\(_2\) [31].

One can also notice that the optical conductivity around InterC is not sensitive to the magnetization or spin splitting in bands, while InterB is very sensitive and appears after magnetic phase transition. Considering the optical selection rule, including inter-transition energy, the enhanced joint density of states from roughly parallel bands and the angular momentum change without spin flipping, we attribute the InterC coming from to the interband transitions marked by blue arrows in Fig. 3a and c. The associated occupied and unoccupied bands are from Co 3d\(_{2g}\) and \(e_g\) orbitals, respectively. These bands are separated due to octahedral crystal-field splitting being irrelevant to the spin-exchange splitting. For the InterB, the interband contributions are marked by green arrows, and they are mainly the transitions among bands of Co 3d orbitals in nearly the same spin channel, which is reasonable since there is nearly non spin flipping and sensitive to magnetization.

In the experimental data, below 100 K, besides InterB and InterC, we observed another absorption peak around 310 cm\(^{-1}\) (InterA in Fig. 1c). However, this peak cannot be resolved in calculated spectra (Fig. 3f). This may come from the overlapping of the low-energy peak with the overestimated Drude response. To resolve this, we established a the tight-binding Hamiltonian from Wan-ner function and performed quite dense k-sampling for integration of inter-band transitions. A small absorption peak can be seen after SOC is considered, which is indicated by the red arrow in Fig. S4 in the supplementary material [21]. This peak is around 350 cm\(^{-1}\), which is consistent with the position of InterA in Fig. 1c. Checking the bands near the Fermi level, we notice that the SOC only triggers gaps along the nodal line except for isolated Weyl points. Thus, we ascribe the InterA to the interband transitions between the inverted bands which are gapped by SOC as marked by red arrow in Fig. 3c. These bands are very close to and compose of Weyl nodes.

In summary, we have carried out comprehensive optical and theoretical investigations on the electronic properties of a magnetic Weyl semimetal candidate Co\(_3\)Sn\(_2\)S\(_2\). Ex-
perimental and theoretical results reveal a moderate electron correlation and show that the increasing exchange splitting renormalize the bands continuously, making the SOC induced gap approaching Fermi level. We have clearly identified the bands contributing to each absorption peak, including the magnetization sensitive and non-sensitive ones. The one from bands close to Weyl nodes is quite promising for potential applications. These results strongly suggest that Weyl nodes can be well controlled by magnetization, which is also identified. These results strongly suggest that Weyl nodes can be well controlled by magnetization, which is quite promising for potential applications.

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