Pressure-induced excitations in the out-of-plane optical response of the nodal-line semimetal ZrSiS

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The anisotropic optical response of the layered, nodal-line semimetal ZrSiS at ambient and high pressure is investigated by frequency-dependent reflectivity measurements for the polarization along and perpendicular to the layers. The highly anisotropic optical conductivity is in very good agreement with results from density functional theory calculations and confirms the anisotropic character of ZrSiS. Whereas the in-plane optical conductivity shows only modest pressure-induced changes, we found strong effects on the out-of-plane optical conductivity spectrum of ZrSiS, with the appearance of two prominent excitations. These pronounced pressure-induced effects can neither be attributed to a structural phase transition according to our single-crystal x-ray diffraction measurements, nor can they be explained by electronic correlation and electron-hole pairing effects, as revealed by theoretical calculations. Our findings are discussed in the context of the recently proposed excitonic insulator phase in ZrSiS.

Topological nodal-line semimetals (NLSMs) with linearly dispersing electronic bands, which cross along a line in reciprocal space, host two-dimensional (2D) Dirac fermions and are currently extensively investigated due to their exotic and highly interesting physical properties [1, 2]. The layered material ZrSiS is considered as the prototype NLSM, where the linearly dispersing bands extend over a large energy range ∼2 eV around the Fermi energy $E_F$, without the presence of topologically trivial bands in the vicinity of $E_F$, and the corresponding nodal lines form a three-dimensional cage-like structure [3]. There are additional Dirac crossings at the X and R point of the Brillouin zone located ∼0.5 eV above and below $E_F$, which are protected by nonsymmorphic symmetry against gapping due to the spin-orbit coupling. The unconventional mass enhancement of quasiparticles in ZrSiS [6] suggests the importance of electronic correlations, which could potentially drive the material toward an excitonic insulator phase or a quantum critical region close to it [7–9].

The exceptional electronic band structure of ZrSiS and related materials ZrXY, where X is a carbon group element (X = Si, Ge, Sn) and Y is a chalcogen element (Y = S, Se, Te) [10], is mainly due to the 2D square nets of Si atoms parallel to the ab-plane, which are the main structural motif besides the square nets of Zr and chalcogen atoms, stacked perpendicular to the ab-plane [see inset of Fig. 1(b)]. Further interesting properties of ZrSiS include high charge carrier mobility and exceptionally large magnetoresistance due to electron-hole symmetry [11–13]. Also the electrodynamic properties of ZrSiS are unusual, with a nearly frequency-independent optical conductivity $\sigma_\text{E}$ for frequencies from 250 to 2500 cm$^{-1}$ (30–300 meV) [14]. This rather flat behavior of $\sigma_\text{E}$ is followed by a U-shaped profile between 3000 and 10.000 cm$^{-1}$ (0.37–1.24 eV), which was ascribed to transitions between the linearly crossing bands of the nodal line close to $E_F$, and a peak located at ∼11.800 cm$^{-1}$ (∼1.46 eV), associated with transitions between parallel bands of the Dirac crossings protected by nonsymmorphic symmetry [15].

All previous experimental studies on the electrodynamic properties of ZrSiS focused on the in-plane optical response, i.e., for the polarization $E$ of the incident electromagnetic radiation aligned along the layers in the ab-plane [see inset of Fig. 1(b)]. In this paper, we report on the out-of-plane optical conductivity of ZrSiS, as obtained by frequency-dependent reflectivity measurements for E directed perpendicular to the layers, i.e., $E_\text{c}$. Furthermore, we studied the in-plane and out-of-plane optical conductivity of ZrSiS under external, quasi-hydrostatic pressure, combined with pressure-dependent single crystal x-ray diffraction (XRD) measurements. In the out-of-plane optical response two new excitations appear under pressure, which cannot be reproduced by density functional theory (DFT) calculations at the generalized gradient approximation [16] level or even by including electronic correlations at GW level and electron-hole pairing effects. Our findings add yet another interesting

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FIG. 1. Optical response functions of ZrSiS at ambient-
conditions for polarization directions E∥ab and E∥c: (a) ref-
lectivity spectra and (b) real part of the optical conductivity, σ1. Inset of (a): loss function -Im(1/ε̂) where ̂ε is the com-
plex dielectric function. Inset of (b): Crystal structure of
ZrSiS with Si square nets parallel to the polarization along (E∥ab).

FIG. 2. Pressure-dependent optical conductivity σ1 with the
Drude-Lorentz fitting and the corresponding contributions at
0 GPa for (a) E∥ab and (b) E∥c. Arrows mark the most pro-
nounced pressure-induced changes in the spectra. Inset of (a):
Comparison between the experimental and both DFT and
BSE calculated interband conductivity σ1,interband at 0 GPa.

facet to the exceptional properties of ZrSIS.

Ambient-pressure reflectivity spectra of ZrSiS for the
polarization along (E∥ab) and perpendicular (E∥c) to the
layers are depicted in Fig. 1(a). (See the Supplemental
Material for a description of sample preparation, ex-
perimental details, and analysis of reflectivity data.) For
both polarization directions, the reflectivity is high at low
energies and shows a distinct plasma edge, indicating the
metallic character consistent with recent resistivity mea-
surements. The anisotropic character of ZrSiS is
manifested by the polarization-dependent energy posi-
tion of the plasma edge, which is shifted towards lower
energies for E∥c compared to E∥ab. Consistently, the
intraband plasmon peak in the loss function defined as
-Im(1/ε̂), where ̂ε is the complex dielectric function, ap-
pears at lower energy, ≈0.47 eV, for E∥c as compared to
≈1.07 eV for E∥ab [inset of Fig. 1(a)]. The anisotropic
optical response is also seen in the real part of the op-
tical conductivity spectrum σ1, displayed in Fig. 1(b).
For both directions, σ1 consists of a Drude term at low
energies due to itinerant charge carriers. From the spec-
tral weight analysis of the Drude contribution we obtain a
plasma frequency ωp of 3.17 eV for E∥ab and 1.08 eV
for E∥c, in agreement with the results of first-principles
calculations. The ratio of dc conductivities σab/σc
amounts to ~16, which lies in between the values 8 and
30 given in Refs. 18 and 19, respectively. Obviously,
the charge dynamics and charge transport is much en-
hanced along the layers compared to the perpendicular
direction, as expected.

Also the profile of the optical conductivity spectrum is
strongly polarization-dependent [Fig. 1(b)], in very good
agreement with the theoretical results of Refs. 18, 21.
For E∥ab, the low-energy σ1 spectrum consists of a Drude
term and a rather flat region up to ~3000 cm−1 followed
by a U-shape frequency dependence, which is bounded
by a rather sharp peak at high frequencies. This
sharp peak (called L4 in the following) is associated
with transitions between parallel bands of the Dirac cross-
ings, which are protected by nonsymmorphic symmetry
against gapping. The profile of the E∥c optical con-
ductivity is markedly different: It is rather featureless,
namely besides the Drude peak it shows only an absorp-
tion band at ~2400 cm−1 and a monotonic increase above
~6000 cm−1, which originates from transitions between
Dirac bands and states further away from EF. Compared
to E∥ab, the out-of-plane momentum matrix elements ex-
hbit substantially weaker k- and band dependence, and
thus the E∥c optical conductivity reflects the behaviour
of the particle-hole (joint) density of states (divided by
frequency) 13. For both directions, the optical conduc-
tivity and reflectivity spectra can be well fitted with the
Drude-Lorentz model (see Fig. 1). The obtained Drude

and Lorentz contributions at ambient pressure are shown in Fig. 3(a) and (b) for $E∥ab$ and $E∥c$, resp.. For comparison with the results of DFT calculations (see the Suppl. Material [17] for details) we subtracted the Drude term from the total $\sigma_1$ spectrum and obtained the contributions from the interband transitions, $\sigma_{1,\text{interband}}$. The interband conductivity spectra agree well with the corresponding theoretical spectra [see inset of Fig. 3(a)].

In the following, the main focus will be on the optical response of ZrSiS under external pressure. The experimental in-plane and out-of-plane $\sigma_1$ spectra are depicted for selected pressures in Figs. 2(a) and (b), respectively. First, we discuss the results for the in-plane optical response. One notices that the induced changes for $E∥ab$ are only modest, and the characteristic profile of the optical conductivity with its U-shape is unchanged up to 7 GPa. A detailed analysis reveals a slight increase of $\sigma_1$ below $\sim$3000 cm$^{-1}$ and a shift of the L4 peak to higher energies with increasing pressure. A comparison between the experimental and theoretical interband optical conductivity from DFT calculations for $E∥ab$ is given in Fig. 3(a) for two selected pressures (2.0 and 7.0 GPa). Like for the experimental results, the U-shape of the theoretical spectrum is retained up to the highest measured pressure and the L4 peak at the high-energy bound of the U-shape is blue-shifted. According to the behavior of the L4 peak, pressure induces a shift of the nonsymmorphic-symmetry protected Dirac crossings away from $E_F$, as a result of the compression of the crystal lattice.

Consistently, the thermal contraction of the crystal lattice during cooling causes a blue shift of the L4 peak [see Fig. 3(c)] [22]. A comparison between the effect of cooling and pressure on the energy position of the L4 peak is given in Fig. 3(d), whereby for the latter both experimental and DFT results are displayed [23]. To conclude, tensile strain, instead of compressive strain, would be needed to push the non-symmorphic symmetry protected Dirac nodes in ZrSiS toward $E_F$, in order to study the expected distinct physics related to these 2D Dirac fermions [21].

Next, we will discuss the pressure-induced effects on the out-of-plane optical conductivity [see Fig. 2(b)]. Starting from the lowest applied pressure (2 GPa), drastic changes occur in the profile of the $E∥c$ $\sigma_1$ spectrum: besides the pressure-induced increase below $\sim$5000 cm$^{-1}$, two new excitations labelled F1 and F2 appear, which gain spectral weight with increasing pressure [25]. Similar to the in-plane optical response, a Drude-Lorentz model was applied for fitting the experimental spectra. As an example, we depict in Fig. 3(b) the experimental interband conductivity $\sigma_{1,\text{interband}}$ at 7 GPa, where the Drude term was subtracted from the total $\sigma_1$, together with the Lorentz contributions. Each of the two new excitations F1 and F2 can be well described by one Lorentzian term. With increasing pressure, the energy position of excitation F2 is almost unchanged, whereas F1 first shifts slightly to lower energies for pressures up to $\sim$4 GPa, and for pressures above 4 GPa this redshift gets more pronounced [Fig. 3(c)]. The oscillator strength of F1 and F2 slightly increases with increasing pressure up to 4 GPa and increases strongly above $\sim$4 GPa [inset of Fig. 3(c)] due to a transfer of spectral weight from the energy range above $\sim$1.9 eV. In Fig. 3(b) we compare the experimental $E∥c$ $\sigma_{1,\text{interband}}$ spectrum for two selected pressures 2.0 and 7.0 GPa with the corresponding DFT results. Interestingly, the theoretical interband conductivity for $E∥c$ is basically unchanged upon pressure application, in strong contrast to the experimental results. In particular, the two excitations F1 and F2 are not reproduced in the pressure-dependent theoretical spectra. Thus, the excitations F1 and F2 should be attributed to effects, which are not included in the band structure calculations, and hence the role of beyond-DFT effects might be relevant, as discussed below.

For an interpretation of our findings, we performed a high-pressure XRD study on a ZrSiS single crystal (see the Suppl. Material [17] for details). With increasing pressure, the lattice parameters $a$ and $c$ monotonically decrease, resulting in a monotonic volume decrease [see Fig. 3(f)]. Further investigation of the reciprocal space up to maximum measured pressure (6.9 GPa) does not reveal the formation of additional or superstructural Bragg reflections [17]. Hence, our diffraction data do not provide any hint for a structural phase transition up to at least 7 GPa, in agreement with Refs. [26, 27]. This finding is in contradiction with results of powder XRD experiments, which suggested a structural phase transition at elevated pressures [28]. We fitted the volume $V$ with the second-order Murnaghan equation of state (EOS) [29] according to $V(p) = V_0 \cdot (B'_0 / B_0) \cdot (p + 1)^{-1} / B'_0$, where $B_0$ is the bulk modulus, $B'_0$ its pressure derivative, which is fixed to 4, and $V_0$ the volume, all at $P=0$ GPa. From the fitting we obtain the value $B_0=144\pm5$ GPa, consistent with earlier reports [28, 31].

Based on the results of our high-pressure XRD study, we can discuss the optical data in more detail. First, it has been proposed that the distance of the nonsymmorphic Dirac crossings from $E_F$ is inversely proportional to the distance between the Si atoms in the Si-Si square nets [31] and hence inversely proportional to the lattice parameter $a$. Accordingly, the energy position of the related L4 peak in the $E∥ab$ $\sigma_1$ spectrum should scale with $1/a$. However, based on our pressure-dependent optical data, we cannot confirm such a behavior. Second, the experimental optical data show an anomaly at $\approx$4 GPa in the shift of the F1 excitation and in the oscillator strength of the F1 and F2 excitations. Since our pressure-dependent XRD results do not provide any evidence for a structural phase transition, this anomaly arises from purely electronic behavior, like in pressurized ZrSiTe [32, 33]. The origin of the excitations F1 and F2, which appear under pressure in the experimental $E∥c$ optical conductivity remains, however, unclear.

It is interesting to note that, based on calculations for a bi-layer square lattice model, Rudenko et al. [7] suggested that ZrSiS undergoes a condensation of interlayer zero-momentum excitons due to electronic corre-
lattices and a high degree of electron-hole symmetry of the electronic band structure, which gives rise to an excitonic insulator state at low temperature. In this weak-coupling scenario (formally similar to BCS superconductivity), a gap opens at $E_F$ in the excitonic insulator state, which leads to a spectral weight transfer and appearance of Hebel-Slichter-like peaks [7, 34]. Transitions between these peaks could in principle lead to distinct excitations of Hebel-Slichter-like peaks [7, 34]. Transitions between the creation of a finite-momentum exciton accompanied by a phonon or another exciton, in order to ensure the momentum conservation. A first step in analysis of such a scenario would be extending the BSE analysis to finite momentum transfer.

Another scenario was proposed recently [8] suggesting that ZrSiS should be located in a quantum critical region between the NLSM and excitonic insulator phases, which could explain the observed quasiparticle mass enhancement [8] in the absence of a pseudogap, consistent with reported photoemission spectra and our optical data. Nevertheless, both the excitonic insulator and quantum critical scenarios are at odds with our observation of pressure-insensitive in-plane response of the L4 peak (e) Pressure-dependent frequency position and oscillator strength (inset) of the peaks F1 and F2. (f) Volume $V$ of the unit cell and lattice parameters $a$ and $b$ as a function of pressure. The solid line is a fit with a Murnaghan-type EOS (see text).

To inspect the role of zero-momentum excitons in the formation of the F1 and F2 peaks we have computed the interband optical spectra by solving the Bethe-Salpeter equation (BSE) with quasiparticle energies calculated at GW level, also testing the impact of the coupling between the resonant and anti-resonant excitations. The results, depicted in the insets of Fig. 2(a) and Figs. 3(a) and (b), show that the optical conductivity is only marginally affected by the electronic correlation and excitonic effects: the BSE $\sigma_1$ spectra are very similar to the DFT spectra and do not exhibit any evident pressure dependence. Formation of a single exciton thus cannot explain our experimental results. A more complex possibility would be the creation of a finite-momentum exciton accompanied by a phonon or another exciton, in order to ensure the momentum conservation. A first step in analysis of such a scenario would be extending the BSE analysis to finite momentum transfer.

In conclusion, according to our reflectivity study, the optical response of the NLSM ZrSiS is highly anisotropic. The polarization-dependent optical conductivity at ambient pressure is in very good agreement with the results of DFT calculations. The in-plane optical response shows only modest changes under pressure, consistent with theoretical predictions. In stark contrast, the out-
of-plane optical conductivity undergoes strong changes under pressure, with the appearance of two pronounced peaks. The observed pressure-induced changes can neither be attributed to a structural phase transition according to our single-crystal XRD data, nor can they be explained by electronic correlation effects and single exciton formation according to our theoretical calculations.

ACKNOWLEDGMENTS

We thank S. Sharma for fruitful discussions. C.A.K. acknowledges financial support from the Deutsche Forschungsgemeinschaft (DFG), Germany, through grant no. KU 1432/13-1. RS acknowledges financial support provided by the Ministry of Science and Technology in Taiwan under project number MOST-108-2112-M-001-049-MY2 and acknowledge for the Academia Sinica for the budget of AS-iMATE-109-13.

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