Searching for a promising topological Dirac nodal-line semimetal by angle resolved photoemission spectroscopy

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Abstract

Topological semimetals, in which conduction and valence bands cross each other at either discrete points or along a closed loop with symmetry protected in the momentum space, exhibited great potential in applications of optical devices as well as heterogeneous catalysts or antiferromagnetic spintronics, especially when the crossing points/lines matches Fermi level ($E_F$). It is intriguing to find the ‘ideal’ topological semimetal material, in which has a band structure with Dirac band-crossing located at $E_F$, without intersected by other extraneous bands. Here, by using angle resolved photoemission spectroscopy, we investigate the band structure of the so-called ‘square-net’ topological material ZrGeS. The Brillouin zone (BZ) mapping shows the Fermi surface of ZrGeS is composed by a diamond-shaped nodal line loop at the center of BZ and small electron-like Fermi pockets around X point. The Dirac nodal line band-crossing located right at $E_F$, and shows clearly the linear Dirac band dispersions within a large energy range $>1.5$ eV below $E_F$, without intersected with other bands. The obtained Fermi velocities and effective masses along Γ–X, Γ–M and M–X high symmetry directions were 4.5–5.9 eV Å and 0–0.50 m_e, revealing an anisotropic electronic property. Our results suggest that ZrGeS, as a promising topological nodal line semimetal, could provide a promising platform to investigate the Dirac-fermions related physics and the applications of topological devising.

1. Introduction

As a novel class of topological matters, topological semimetals (TSM) own unique electronic band structures, i.e. the conduction and valence bands touch at certain points or lines near Fermi level ($E_F$) in the Brillouin zone (BZ), corresponding to topological nodal-point (Weyl-type or Dirac-type)/nodal-line semimetals, respectively [1, 2]. The electronic excitations around the band-crossing points in TSM can be analogous to the relativistic Weyl or Dirac fermions, making it an important platform to investigate fundamental high-energy physics in condensed matter systems [3]. Besides, owing to the outstanding electronic, optical, thermal and magnetic properties, TSM can be broadly applied in some important areas, such as topological quantum computation [4], photovoltaic devices [5], thermoelectric [6] and magneto thermoelectric [7] applications, catalysis [8] and so on. Thus, the prediction [9, 10], material preparation and characterization [11, 12] of TSM have attracted much attentions in recent years.

Most applications require that the Dirac points (DP) or nodes match $E_F$, accompanied with zero effective mass [13]. For example, as a promising route toward topological quantum computation, Majorana excitations (whose antiparticle is itself) are much concerned [14], and it requires mandatorily that the Dirac...
Figure 1. Crystalline structure of ZrGeS. (a) The ball-and-stick model of crystalline structure of ZrGeS. The arrow indicates that the natural cleavage plane between adjacent Zr-S layers, whose interlayer coupling is relatively weak. (b) Single crystal x-ray diffraction pattern for ZrGeS.

point matches $E_F$ rather exactly in order to stabilize and separate it from the conventionally confined states within vortices [4]. In metal–semimetal–metal terahertz device, owing to the excitation of Dirac fermions, when the Dirac node is tuned to $E_F$, it shows colossal photoresponse improvement at terahertz frequency, with a room-temperature photoresponsivity of 0.52 A W$^{-1}$ at 0.12 THz and 0.45 A W$^{-1}$ at 0.3 THz [5]. In magneto-thermoelectric application, Nernst coefficients increases progressively as $E_F$ approaches the Dirac point and reach the maximum when they matched, exhibiting an even function at series of carrier densities and temperatures [7]. In addition, some important chemistry reactions are favored when the Dirac point meet $E_F$, such as Diels–Alder reaction [15]. Besides, it is also benefit to study the topological property and classification when the Dirac point locate around $E_F$ [16], avoid the influence of other non-topological bands [17].

However, due to global charge cancellation, the point nodes of band crossing usually does not prefer to locate at $E_F$ [18]. Hence, it is important to find a material with Dirac point matches $E_F$ and investigate its electronic band structure. Here, we report the experimental investigations on the band structure and Fermi surfaces (FS) of topological Dirac nodal-line semimetal [3] ZrGeS by angle resolved photoemission spectroscopy (ARPES). Our results show that the FS of ZrGeS is composed by a diamond-shaped nodal line loop and small electron-like Fermi pockets around X point. Within $>1.5$ eV large energy range below $E_F$, the nodal line shows linear Dirac band dispersions without intersected by other extraneous bands. Moreover, the crossing Dirac point of nodal line is located rightly at $E_F$. In addition, the band structure around X point, the Fermi velocities and effective masses along $\Gamma$–X, $\Gamma$–M and M–X high symmetry directions were obtained, suggesting an anisotropic electronic property.

2. Experimental methods

The ZrGeS single crystals (figure 1(a)) were synthesized by a chemical vapor transport method [19]. The ARPES measurements were performed at the vacuum interconnected nanotech workstation of SINANO with a ScientaOmicron DA30L analyzer and monochromatized He I$\alpha$ ($h\nu = 21.218$ eV) light source. The samples were cleaved in situ and measured at 81 K with a background vacuum better than $5 \times 10^{-11}$ mbar. The cleaved crystal was checked by single crystal x-ray diffraction (Rigaku Rapid IIR).

3. Results and discussion

As shown in figure 1(a), ZrGeS compound belongs to a large family of square-net materials with the nonsymmorphic PbFCl-type structure ($P4/nmm$, no. 129) [1], and possesses a tetragonal layered structure with the Ge square-net plane, sandwiched by the Zr–S layers, forming the S–Zr–Ge–Zr–S slabs. The interlayer coupling between the adjacent Zr–S layers is relatively weak, which provides a natural cleavage plane in between [19] and yields a (001) surface with S termination, similar to its isostructural compound ZrGeSe [20]. The experimental lattice constants are $a = b = 3.63$ Å and $c = 8.02$ Å [21]. After cleavage, the high quality of crystallinity is verified by the sharp and clean (00L) x-ray diffraction peaks, as shown in figure 1(b), informing a (001) surface obtained.
Figure 2(a) shows the surface projected BZ of ZrGeS, with high-symmetry points indicated. The measured Fermi surface map is shown in figure 2(b), consisting of the diamond-shaped nodal line loop FS around the BZ center (Γ') point (also see figure 3 and the illustrated nodal line in figure 2(a)) and small electron-like Fermi pockets around the X point (also refer to figure 4). Also, the constant energy maps (CEMs) at −0.222 eV, −0.432 eV and −0.642 eV were measured (figures 2(c)–(e)). As $E - E_F$ decrease from 0 eV to −0.642 eV, single nodal line state gradually split into two bands (also see figure 3), indicating hole-like Fermi pockets. For the elliptic-like state around X point, it shrinks first with crossed at −0.432 eV and then expands, indicating a linear Dirac band with Dirac point at −0.432 eV (also refer to figure 4), similar to the previously established electronic structures of ZrGeSe [20, 22], ZrGeTe [22, 23], ZrSiS [24, 25], ZrSiSe [25], ZrSiTe [25] and ZrSnTe [26, 27].

Figure 3 shows the band dispersions measured along the Γ–M direction and parallel to Γ–M direction with a 0.26 (1/Å) shifting. The similarity between figures 3(a) and (b) indicates the nodal-line structure, which agrees very well with the nodal line loop FS observed in figure 2(b) and theoretical calculation [1, 28]. In the large energy range from $E_F$ to −1.5 eV, a linear Dirac-like dispersion without other extraneous bands was observed. By measuring the slope of the linear bands, we obtained the Fermi velocity $\hbar v_F \sim 5.4$ eV Å, just slightly lower than that of graphene ($\hbar v_F \sim 6.7$ eV Å) [29]. We are aware of the crossing Dirac point 1 (labeled as DP1) located at $E_F$, both from the dispersion map (figures 3(a)–(b)) and momentum distribution curve (MDC) plot (figure 3(c)). In order to measure the position of DP1 more accurately, Voigt function were used to fit [4] the MDCs. As shown in figure 3(d), while the MDC at -0.3 eV can be fitted into two peaks, the MDC at $E_F$ can only be fitted by one peak, suggesting that DP1 locates at $E_F$ accurately, more consistent with the calculated result (i.e. located almost at $E_F$) [30, 31] than that roughly estimated to be >0.2 eV above $E_F$ by extrapolating the bands parallel to Γ–M direction, reported by Nakamura et al [22]. According to the reported ARPES measurements of ZrHM ($H = Si/Ge, M = S/Se/Te$) family materials, the correspond Dirac point along Γ–M direction of ZrSiS [32] located below $E_F$, while ZrSiSe [25] located above $E_F$ but lower than ZrSiTe [25], revealing the dependency that the locations of Dirac point increase as $M$ element changing from S to Te, in consistence with the theoretical calculations [30]. Then, it is reasonable that the Dirac point of ZrGeS here locate at $E_F$, lower than ZrGeSe [20] and ZrGeTe [23]. Note the correspond Dirac point location estimated from Nakamura et al (>0.2 eV) [22] is slightly higher than that of ZrGeSe (0.182 eV) [20], our result here shows better coincident with prediction. This difference may benefit from the high single crystal quality of our ZrGeS sample, as the XRD result shown in figure 1(b). In addition, Schoop et al predicted that for the square-nets materials, the exact position of $E_F$ is determined by the number of electrons in square-net (such as Ge net in ZrGeS), and
it will match the nodal point when an electron count is six electrons per net atom [1, 33]. Based on this assumption, each Ge atom will own six electrons, then the valence state of Ge will be $-2$, resulted in half-filled $p$ and $p_y$ orbitals.

Following, we discuss the spin–orbit coupling (SOC) induced gap [25] in ZrGeS. For ZrHM family materials, the Dirac nodal line along $\Gamma$–M (figures 2 and 3) and nodal point along $\Gamma$–X (figure 4) directions are protected by the symmorphic $C_{2v}$ symmetry, while the Dirac point at X (figure 4) is protected by the non-symmorphic symmetry. With SOC, the $C_{2v}$ point group allows only one irreducible representation, and therefore gaps out the $\Gamma$–X and $\Gamma$–M crossings, while the non-symmorphic protected crossings at X is robust against SOC [1, 34, 35]. On the other hand, the size of the SOC induced gap is approximately proportional to the fourth power of the nuclear charge $Z$ [36]. Then the gap in ZrGeS should be larger than ZrSiS but smaller than ZrGeSe, as the nuclear charge of Ge is larger than Si while the one of S is smaller than Se. According to the reported theoretical calculations, the SOC induced gaps in ZrSiS [35] and ZrGeSe [22] are $\sim$20 meV and 30 meV, respectively. Then the SOC induced gap in ZrGeS will be 20–30 meV, which is small, similar to ZrSiS [25], and consistent with the quantum oscillation study of ZrGeS [19]. Furthermore, the opening of the SOC-induced gap normally leads the energy band to deviate from linear dispersion [19], but no obvious deviation was observed in our results, also suggesting the SOC induced gap.
Figure 4. Band structure along $\Gamma$–X and M–X directions around X point. (a) and (b) are the measured dispersion maps along $\Gamma$–X and M–X directions, respectively. The dashed lines indicate the linear Dirac bands, ‘DP2’ and ‘DP3’ indicate the DP, ‘SS’ represent for SS. (c) and (d) The correspond MDC plots for the energy range from $E_F$ to $-1.0$ eV in (a) and (b), the momentum positions of SS and linear Dirac bands are indicated by triangles and dashed lines, respectively.

here is small, although it is much larger than that of graphene (24 $\mu$eV) [37], a kind of Dirac semimetal with Dirac cone point located at $E_F$ [16, 38, 39].

For ZrGeS’s isostructural compound ZrSiTe, in which SOC can induce a larger gap (100 meV) [41] than ZrGeS (20–30 meV) along $\Gamma$–M nodal line, but the topological nature of the nodal line is still remained in ZrSiTe [40]. Then, the nodal line (DP1) in ZrGeS should be topological similarly. In fact, previous quantum oscillation study of ZrGeS also revealed that ZrGeS is a kind of topological semimetal [19]. Besides, the tolerance factor ($t$), defined as the ratio of the distance of atoms in the square net and the distance to the nearest neighboring atom of an adjacent layer in the structure, turned out to be a powerful tool to predict topological semimetals and the results suggest it will be topological semimetal when $t \leq 0.95$ [28]. Here, we calculate the tolerance factor $t$ of ZrGeS to be 0.896 according to ZrGeS’s crystal structure, smaller than the critical value 0.95, suggesting ZrGeS as a topological semimetal. Briefly, ZrGeS has rather simple topological Dirac nodal-line bands within a large energy range $>1.5$ eV without intersected by other extraneous bands, and the Dirac point is located at Fermi level ($E_F$). All these results together suggest that ZrGeS is potentially a promising topological nodal line semimetal (TNLSM) [3] without SOC. Considering a small gap opened with SOC, further theoretical and experimental research is needed in order to realize ideal nodal line semimetal [3, 41] robust against SOC in real material. It is worth mentioning that, negative longitudinal
magneto-resistance (LMR) was observed in the quantum oscillation study of ZrGeS [19], and it is most possibly related with chiral anomaly [42, 43], as ZrGeS own relatively low mobility (<1700 cm² (Vs)⁻¹) [19] refer to NaBi (2600 cm² (Vs)⁻¹), in which typical chiral anomaly induced LMR was evidenced [43]. In addition, ZrGeS owns non-zero Berry phase and Zeeman splitting [19], implying that anomalous Hall effect [44–46] may possibly be observed in ZrGeS. Moreover, large spin Hall effect has been reported in ZrGeS [47]. These above together also support that ZrGeS is a kind of promising topological semimetal.

In addition, we investigate the electronic band structures of ZrGeS around X point (figure 4). Again, in the large energy range from $E_F$ to $-1.5$ eV, the linear Dirac bands were observed, intersecting with the surface state (SS) along $\Gamma$–X direction, in consistence with theoretical prediction [21, 30, 31] and similar to the results of other ZrGeS’s isostructural compounds [23, 27, 48]. By measuring the slope of these linear bands, we find the Fermi velocity of the inner (labeled with black dashed lines), outer bands (labeled with green dashed lines) along $\Gamma$–X and the bands along X-M direction to be $\hbar v_F \sim 4.5$ eV Å, 4.9 eV Å and 5.9 eV Å, respectively, similar to its isostructural compound ZrGeSe [20].

On the other hand, we observe these inner Dirac bands cross at nonsymmorphic-symmetry protected four-fold degeneracy X point at $-342$ meV (Dirac point 2, labeled as DP2) (figures 4(a) and (c)). The Dirac point of outer band locates partially above $E_F$, which is invisible by standard ARPES measurement, similar to the results of other $ZrHM$ ($H = Si/Ge, M = Si/Se/Te$) materials [20, 22, 23, 27, 32, 35], as predicted by theoretical calculations [21, 30, 31]. By extrapolating the outer linear bands, the Dirac point is estimated to be located at 519 meV. Moreover, we aware that the location of DP2 in ZrGeS ($-0.43$ eV) here is similar to ZrGeSe ($-0.48$ eV) [20] and ZrGeTe ($-0.40$ eV) [23], but lower than ZrSiSe ($-0.40$ eV) [25] and ZrSiTe ($-0.30$ eV) [25, 31]. Note the crystallographic $c/a$ ratio of ZrGeS (2.212) is similar to ZrGeSe (2.232) and ZrGeTe (2.224), but smaller than ZrSiSe (2.309) and ZrSiTe (2.573) [32, 49]. It seems that the position of DP2 move closer to $E_F$ as the $c/a$ ratio increase, in consistence with the findings of Andreas et al [49].

Figures 4(b) and (d) also show the predicted Dirac-like surface states [21, 30, 31] along the $M$–X–M high-symmetry direction with the Fermi velocity $\hbar v_F \sim 5.9$ eV Å and the Dirac point DP3 located at $-342$ meV. The DP3 here is located closer to $E_F$ than that of along $\Gamma$–X direction (DP2, $-342$ meV), similar to other isostructural compounds [20, 23–25, 32, 35, 42]. According to theoretical calculations [21, 30, 31] and photon energy dependent ARPES measurements of ZrSiS, ZrGeTe [23] and ZrSnTe [27], the linear Dirac bands along $\Gamma$–X and X–M directions are originated from bulk and surface states, respectively, which cross at X point with different energies. Besides, the Fermi wave vector along X–M direction $k_{\Gamma X, inner} = 0.100 (1/Å)$ (figure 4(a)) is smaller than the inner bands along $\Gamma$–X direction $k_{\Gamma X, inner} = 0.038 (1/Å)$ (figure 2(a)), in consistence with the elliptical FS property around X point (figure 2(a)).

At last, we could extract the effective mass $m^*$ from our experiments. For the linear Dirac bands, analogous to the relativistic Dirac fermions [3, 11], the effective mass $m^*$ can be evaluated by ARPES through Fermi velocity $\hbar v_F$ and Fermi wave vector $k_F$ by the formula $m^* = \hbar k_F / v_F$ [39]. Inputting with $\hbar v_F$ and $k_F$ of the linear bands along inner and outer bands, $\Gamma$–X and $\Gamma$–M directions are 4.5, 4.9, 5.9, 5.4 eV Å and 0.100, 0.318, 0.038, 0 (1/Å), respectively, here we obtain $m^*_{\Gamma X, inner} = 0.17 m_e$, $m^*_{\Gamma X, outer} = 0.50 m_e$, $m^*_{X M} = 0.05 m_e$ and $m^*_{\Gamma M} = 0 m_e$, where the subscript ‘inner’ and ‘outer’ refer to the inner and outer bands along $\Gamma$–X direction in figure 4(a) and $m_e$ is the free electron mass. We note that the values of $m^*$ along different high-symmetry directions are different, revealing an anisotropic electronic property. Besides, owing to the fact that Dirac point of the linear bands along $\Gamma$–M direction matches $E_F$, the corresponding $k_F$ is zero, which results in the zero effective mass, similar to the situation of graphene [13].

4. Conclusion

The band structure and FS topology of TNLSM ZrGeS was investigated systematically by ARPES. We find that the FS is composed by a diamond-shaped nodal line loop around $\Gamma$ point and small electron-like Fermi pockets around X point. Dirac-like linear bands were observed along $\Gamma$–M, $\Gamma$–X, and M–X directions, within a large energy range (>$1.5$ eV). Specially, the DP of the bands along and parallel to $\Gamma$–M directions locate at Fermi level, without intersected by other extraneous bands, suggesting ZrGeS to be a kind of promising topological Dirac nodal line semimetal. Besides, the obtained Fermi velocities and effective masses along $\Gamma$–X, $\Gamma$–M and M–X high-symmetry directions are different, ranged from 4.5 eV Å to 5.9 eV Å and from 0 $m_e$ to 0.50 $m_e$, respectively, suggesting an anisotropic electronic property.
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Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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