Bulk superconductivity in the Dirac semimetal TlSb

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A feasible strategy for realizing the Majorana fermions is searching for a simple compound with both bulk superconductivity and Dirac surface states. In this paper, we perform calculations of electronic band structure, the Fermi surface, and the surface states, and measure the resistivity, magnetization, and specific heat of a TlSb compound with a CsCl-type structure. The band structure calculations show that TlSb is a Dirac semimetal when spin-orbit coupling is considered. TlSb is first determined to be a type-II superconductor with $T_c=4.38$ K, $H_{c1}(0)=148$ Oe, $H_{c2}(0)=1.12$ T, and $\kappa_{GL}=10.6$. We also confirm that TlSb is a moderately coupled $s$-wave superconductor. Although we cannot determine the band near the Fermi level $E_F$ that is responsible for superconductivity, its coexistence with topological surface states implies that the TlSb compound may be a simple material platform to realize the fault-tolerant quantum computations.

$s$-wave superconductor, Dirac semimetal, topological superconductor

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1 Introduction

Topological superconductors host Majorana fermions, described by a real wave function, which provide protection for quantum computations [1]. Realization of topological superconductivity (TSC) has become one of the most interesting topics in the condensed matter physics in recent decades. According to the discussion in ref. [2], there are intrinsic and artificially engineered topological superconductors. For the intrinsic type, the topological nontrivial gap function naturally appears. Sr$_2$RuO$_4$ [3-6] was the first proposed topological superconductor although the existence of chiral $p$-wave superconductivity (SC) is still under debate. Cu$_3$Bi$_2$Se$_3$ [7] was the first topological insulator (TI) to show SC ($T_c\sim4$ K) upon doping with charge carriers. TIs are promising materials for two dimensional (2D) TSC due to the survival of topological surface states even when doped with carriers. Many experiments, such as conductance spectroscopy [8], nuclear magnetic resonance (NMR) measurements of the Knight-shift [9], and specific heat in applied magnetic fields [10], have already provided evidence for TSC emerging in Cu$_3$Bi$_2$Se$_3$. Nematic SC discovered in Cu$_3$Bi$_2$Se$_3$ [9, 10] was also observed in similar superconductors derived from Bi$_2$Se$_3$, such

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as in Sr$_2$Bi$_2$Se$_3$ [11], and Nb$_x$Bi$_2$Se$_3$ [12]. Sn$_{1-x}$In$_x$Te [13] is another superconductor formed upon doping charge carriers into a topological crystalline insulator. In the cleanest sample (x~0.04) with the lowest $T_c$ (1.2 K), a pronounced zero-bias conductance peak (ZBCP), similar to that in Cu$_2$Bi$_2$Se$_3$, has been observed by point contact spectroscopy [13]. Another type is the artificially engineered TSC in hybrid structures. According to the idea proposed by Fu and Kane [14], if $s$-wave pairing is imposed on the topological surface states of a three dimensional (3D) TI through the superconducting proximity effect, the resulting superconducting state should be a 2D $p$-wave SC that harbors a Majorana zero mode in the vortex core. Experimentally, proximity-induced SC on the surface of 3D TIs has been studied by many groups [15-21]. The observation [22] of 4$\pi$-period Josephson supercurrent in 3D HgTe TI is encouraging, although, it is difficult to elucidate the topological nature of the induced 2D SC.

There are also controversies regarding emerging TSC and the observed ZBCP being a Majorana zero-energy mode (MZM) in doped topological materials. Realization of TSC through the superconducting proximity effect in hybrid structures has many engineering challenges. A feasible way to achieve such TSC is to realize TSC in a simple compound wherein both the topological surface state and the bulk SC co-exist. Then, Majorana fermions emerge at the edge of the superconductor. Recently, the observations of MZM in the core of a vortex [23, 24], at the end of an atomic defect line [25], near the Bi islands [26], and near interstitial Fe atoms [27] in the simplest Fe-based superconductor Fe$_{1+y}$Te$_{0.5}$Se$_{0.5}$ ($T_c$ = 14 K) [28] have motivated searches for similar systems. TlSb crystallizes in a cubic CsCl structure with space group $Pm\bar{3}m$ (No. 221) (as shown in the inset of Figure 1), and from this structure, a large number of topological semimetal/metals (TMs) have been designed [29]. These TMs range from triple nodal points, type-I nodal lines, and critical type nodal lines, to hybrid nodal lines. For example, CaTe is a typical type-I nodal line and Dirac TM [30]. YIr is a typical triple-nodal-point TM [29] and YMg possesses multiple types of band crossing [29]. Therefore, we attempted to grow TlSb crystals to study its topological nature and SC. Unfortunately, only polycrystalline TlSb samples were obtained, but several interesting properties were still observed.

In this work, we performed calculations of the electronic band structure, the Fermi surface, and the surface states on the (001) plane of TlSb and measured resistivity, magnetization and specific heat of a polycrystalline TlSb sample. The band structure calculations show TlSb is a Dirac semimetal with 4-fold degenerate nodes at $\Gamma$ and R points. It was also found that TlSb is a type-II superconductor with a superconducting transition temperature $T_c$ = 4.38 K, a lower critical field $H_{c1}(0) = 148$ Oe, an upper critical field $H_{c2}(0) = 1.12$ T, and a Ginzburg-Landau (GL) parameter $\kappa_{GL} = 10.6$. The obtained specific heat jump, $\Delta C_{cal}/\gamma_0T_c \sim 1.42$, indicates that TlSb is a conventional phonon-mediated superconductor with $s$-wave superconducting symmetry. These results indicate that both $s$-wave SC and surface states coexist in TlSb. Evidence of Majorana fermions on the edges is to be confirmed in future studies.

**2 Experimental**

Polycrystalline TlSb samples were synthesized by a peritectic reaction method. A mixture of stoichiometric high-purity Tl (99.99%) and Sb (99.999%) powder was placed in an alumina crucible, sealed in an evacuated quartz tube, heated at 650°C for 20 h (until the mixture was completely melted), and then the temperature was decreased to 195°C. To avoid decomposition of the TlSb phase at 191°C [31], the quartz tube was quenched to room temperature at 195°C, however, the TlSb samples always contained a small amount of unreacted Sb impurities due to precipitation of Sb before the peritectic reaction. The obtained TlSb alloy was easy to cut for subsequent structural characterization and property measurements. Polycrystalline X-ray diffraction (XRD) was carried out on a PANalytical diffractometer equipped with CuKα radiation. The TlSb XRD pattern is shown in Figure 1. The primary diffraction peaks were fit by the CsCl-type structure with space group $Pm\bar{3}m$. The lattice parameters $a = b = c = 3.86(5)$ Å were obtained by Rietveld refinement using a generalized structure analysis system (GSAS) [32]. A rectangular bar of the sample was cut for magnetization and resistivity measurements, which were performed on magnetic (Quantum Design, MPMS-7 T) and physical (Quantum Design,
The band structure was calculated using density function theory (DFT) with the WIEN2k package [33]. The generalized gradient approximation (GGA) of the Perdew-Burke-Ernzerhof (PBE) exchange-correlation function [34] was employed for exchange-correlation potential calculations. A cutoff energy of 520 eV and a $13 \times 15 \times 15$ $k$-point mesh were used to perform the bulk calculations. The Fermi surface (FS) was performed with the WannierTools [35] package, which is based on the maximally localized Wannier function tight-binding model [36-38] constructed using the Wannier90 [39] package.

3 Results and discussion

We first discuss the electronic band structure without considering spin-orbit coupling (SOC). As shown in Figure 2(a), both conduction (red) and valence (blue) bands cross the Fermi level $E_F$. There are three bands crossing, with a threefold degenerate at 0.8 and 2 eV below $E_F$ at the high symmetry $\Gamma$ and $R$ points, respectively. However, when SOC is considered (see Figure 2(b)), gaps open and leave two twofold degenerate bands at both high symmetry points. Because both time-reversal and inversion symmetries are present, no spin-splitting occurs and the twofold degenerated bands come

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**Figure 2** (Color online) The electronic band structures of TlSb without (a) and with (b) SOC. (c) The first Brillouin zone and the projected surface Brillouin zone of the (001) plane with high symmetry points. (d) 3D bulk Fermi surfaces and color-coded Fermi velocities (red is high velocity). (e) Calculated Fermi surfaces cross section at the $k_z = 0$ plane. (f) Calculated energy dispersion in the $k_x$-$k_y$ plane at Dirac point located at $R$ point. The $R$ point is highlighted as a pink dot. Note that $k_x = k_y = \pi/a$, and their units are Å$^{-1}$. 

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together to fourfold degenerate points, indicating TlSb is a Dirac semimetal. We also calculated the density of states (DOS), as shown in the right panel of Figure 2(b). The DOS at $E_F$ is mainly the contribution of Sb orbits. To further clarify the band structure of TlSb, we calculated its 3D bulk FS of the first Brillouin zone (BZ), as shown in Figure 2(d), which exhibits complex 3D characteristics. Figure 2(e) presents the FS on the $k_z=0$ plane, which is the cross section passing the $\Gamma$ point of the 3D FS. Figure 2(f) displays the energy dispersion in the $k_x$-$k_y$ plane, in which the Dirac dispersion at the R point (pink dot) is clearly seen, further demonstrating that TlSb is a Dirac semimetal.

We then calculated the surface states on the (001) plane using a surface Green’s function method [40]. As shown in Figure 3(a), the projected Dirac points are hidden in the continuous bulk states; the surface states are shown as red curves. The (001)-surface energy contour is shown in Figure 3(b)-(d) with $E=E_F$, $E=-1.5$ eV and $E=-4$ eV, respectively. The surface band at $E=-4$ eV, deep below $E_F$, can be ignored due to its negligible contribution to the electronic properties of the material. Due to TlSb having inversion-symmetry, its topology can be described by one strong topological index $v_0$ and three weak indices $v_1$, $v_2$, $v_3$ [41]. Thus, we calculated the Wilson loops on six time-reversal invariant planes using WannierTools [39]. The results are shown in Figure 4. According to the definition of Wilson loops [42, 43], the topological indices are $v_0(1;000)$, indicating that TlSb is a strong topological material.

Next, we focus on SC emerging in the Dirac semimetal TlSb. Figure 5(a) displays the temperature dependence of resistivity, $\rho(T)$, measured at zero field. With decreasing temperature, $\rho_{xx}$ slowly decreases, exhibiting poor metallic behavior, then drops to zero at 4.32 K. A superconductivity transition occurs with a mid-temperature $T_{c}^{mid}=4.38$ K, and a transition width $\Delta T_{c}=0.15$ K. The resistivity data above 50 K can be described by the model proposed by Wiesmann et al. [44] with the following formula:

$$\frac{1}{\rho(T)} = \frac{1}{\rho_{\text{max}}} + \frac{1}{\rho_{1} + \rho_{\text{BG}}} ,$$

where $\rho_{\text{max}}$ is the saturating resistivity [45] resulting from Ioffe-Regel scattering, which is typically observed in many

Figure 3 (Color online) (a) Surface band structure for the (001) plane along projected high symmetry points. The surface spectra of the (001) plane with (b) $E=E_F$, (c) $E=-1.5$ eV and (d) $E=-4$ eV.
transition metals or transition-metal compounds, such as A15 compounds Nb$_3$Sb [46] and Nb$_3$Sn [47]. $\rho_1$ is a fitting parameter, and $\rho_{BG}$ is the electron-phonon scattering term described by the Bloch-Grüneisen formula:

$$\rho_{BG}(T) = 4A \left( \frac{T}{\Theta_D} \right)^5 \int_0^{\Theta_D/T} \frac{x^5}{(e^x - 1)(1 - e^x)} dx,$$

where $A$ is a fitting parameter and $\Theta_D$ is the Debye temperature. The fit is shown by the blue curve in Figure 5(a) and yields $\rho_{\text{max}} = 115.88 \, \mu\Omega \, \text{cm}$, $\rho_1 = 61.18 \, \mu\Omega \, \text{cm}$, and the Debye temperature $\Theta_D = 133.1 \, \text{K}$. It was also found that the low temperature resistivity data ($< 20 \, \text{K}$) can be fit by the Fermi liquid behavior $\rho(T) = \rho_0 + AT^2$ with $\rho_0 = 28.91 \, \mu\Omega \, \text{cm}$ and $A = 1.4 \times 10^{-2} \, \mu\Omega \, \text{cm} \, \text{K}^{-2}$ as shown in the inset of Figure 5(a).

The superconducting transition is also confirmed by the susceptibility measurement. Figure 5(b) presents the temperature dependence of susceptibility, $\chi(T)$, measured at $H = 5 \, \text{Oe}$ with both zero-field cooling (ZFC) and field cooling (FC) processes. It is clear that a sharp diamagnetic transition emerges at $4.3 \, \text{K}$. Complete diamagnetism ($4\pi\chi \sim -1$) below $T_c$ indicates bulk superconductivity from TlSb because Sb is non-superconducting at ambient pressure.

Figure 6(a) shows the field dependence of magnetization, $M(H)$, measured at $2 \, \text{K}$ for a TlSb sample. Hysteresis is present, which indicates that TlSb is a type-II superconductor. $M(H)$ was then measured at various temperatures below 4.5 K, as shown in Figure 6(b). The lower critical field $H_{c1}(T)$ can be estimated by the field at which the $M(H)$ curve starts to deviate from linearity. The obtained $H_{c1}(T)$ is shown in the inset of Figure 6(b). The lower critical field at zero temperature $H_{c1}(0) = 148 \, \text{Oe}$ was obtained by fitting using the GL relationship: $H_{c1}(T) = H_{c1}(0)[1 - (T/T_c)^2]$.

In order to get the upper critical field $H_{c2}(0)$, we measured the superconducting transition temperature ($T_{c\text{ind}}$) at various applied magnetic fields. As shown in the inset of Figure 7, the $T_c$ decreases and the transition width $\Delta T_c$ increases with increasing magnetic field. Using the GL formula $H_{c2}(T) = H_{c2}(0)(1 - t^2)/(1 + t^2)$, where $t$ is the reduced temperature $t = T/T_c$, to fit the $H_{c2}(T)$ data, the zero temperature upper critical field $H_{c2}(0) = 1.12 \, \text{T}$ was obtained. This value is much lower than the Pauli limit field $H_{c2}^{\text{Pauli}}(0) = 1.86T_c = 8.18 \, \text{T}$. The GL coherence length $\xi_{\text{GL}}(0) = 15.3 \, \text{nm}$ was then estimated using the formula $H_{c2}(0) = \Phi_0/(2\pi\xi_{\text{GL}}^2)$, where $\Phi_0$ is the quantum flux ($h/2e$). The penetration depth $\lambda_{\text{GL}}(0) = 162 \, \text{nm}$ was estimated using the formula $H_{c1}(0) = (\Phi_0/(4\pi\lambda_{\text{GL}}^2(0)))(\ln(\lambda_{\text{GL}}(0)/\xi_{\text{GL}}(0)))$, and the GL parameter $\kappa_{\text{GL}} = \lambda_{\text{GL}}(0)/\xi_{\text{GL}}(0) = 10.6$.

We also measured the specific heat as a function of temperature, $C_p(T)$, for TlSb in the temperature range of 0.5-5 K at both zero field and 3 T, as shown in Figure 8. It is

Figure 4  Wilson loops of six time-reversal invariant planes at (a) $k_1 = 0.0$, (b) $k_1 = 0.5$, (c) $k_2 = 0.0$, (d) $k_2 = 0.5$, (e) $k_3 = 0.0$, and (f) $k_3 = 0.5$, where $k_1$, $k_2$, and $k_3$ are in units of reciprocal lattice vectors.

Figure 5  (Color online) (a) The temperature dependence of resistivity, $\rho(T)$, and the fit using eq. (1). Inset: the $\rho(T)$ data below 20 K, showing a superconducting transition and $T^2$ behavior. (b) The temperature dependence of magnetic susceptibility, $\chi(T)$, measured at $H = 5 \, \text{Oe}$. 

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clear that the zero-field $C_p(T)$, compared with the $C_p(T)$ measured at 3 T (> $H_{c2}$, in this case bulk SC is completely suppressed), exhibits a small and broad peak near $T_c$, corresponding to the superconducting transition. No other anomaly was observed except for the peak near $T_c$ = 4.38 K. This indicates that no Tl impurities ($T_c$ = 2.39 K) emerged in our sample, although a small amount of non-superconducting Sb impurities was detected in XRD. We fit the low temperature $C_p(T)$ data measured at 3 T using the Debye model:

$$C_p/T = \gamma_n + \beta_1 T^2 + \beta_3 T^4,$$

(3)

where $\gamma_n$ is the Sommerfeld coefficient and both the $\beta_3 T^3$ and $\beta_5 T^5$ are the phonon contributions to specific heat. The parameters $\gamma_n = 5.56$ mJ mol$^{-1}$ K$^{-2}$, $\beta_3 = 1.39$ mJ mol$^{-1}$ K$^{-4}$, and $\beta_5 = 0.44$ mJ mol$^{-1}$ K$^{-6}$ were obtained. The inset of Figure 8 shows the normalized $\Delta C/T = \Delta C/T_c(0) = C_p(T)/C_p(0)$ as a function of the normalized temperature $t = T/T_c$. The bulk superconducting temperature $T_c = 4.3$ K was estimated by an entropy-balance method, which is consistent with the results from the resistivity and susceptibility measurements mentioned above. The normalized specific heat jump $\Delta C/T_c = 1.42$ was estimated and was almost the same as the predicted value (1.43) by the Bardeen-Cooper-Schrieffer (BCS) theory [48]. This indicates that TlSb is a s-wave phonon-mediated superconductor. The Debye temperature $\Theta_D = 141$ K was estimated using the formula $\Theta_D = (12\pi^2 NR/5\beta_3)^{1/3}$, where $N = 2$ is the number of atoms in an unit cell and $R = 8.314$ J mol$^{-1}$ K$^{-1}$ is the molar gas constant, close to that obtained from the $\rho(T)$ data mentioned above. Using the obtained $\Theta_D$ and $T_c$ values, we calculated the electron-phonon coupling constant $\lambda_{ep}$ using the McMillan formula [49]:

$$\lambda_{ep} = \frac{1.04 + \mu^* \ln(\Theta_D/1.45T_c)}{(1 - 0.62\mu^* \ln(\Theta_D/1.45T_c) - 1.04)},$$

(4)

where $\mu^* = 0.13$ is a typical value of the Coulomb repulsion pseudopotential for intermetallic superconductors. The
obtained $\lambda_{ep} = 0.78$ value is comparable to that of other superconductors such as PbTaSe$_2$ ($\lambda_{ep} = 0.74$) [50] and Nb$_{0.18}$Re$_{0.82}$ ($\lambda_{ep} = 0.73$) [51], suggesting TISb is a moderately coupled superconductor. The obtained superconducting parameters are summarized in Table 1.

4 Conclusion

In summary, calculations of the electronic band structure, the FS, and surface states show that TISb with a CsCl-type structure is a Dirac semimetal. We measured the resistivity, magnetization, and specific heat for a polycrystalline TISb sample. We first found that TISb is a type-II superconductor with $T_C = 4.38$ K, $H_{c1}(0)=148$ Oe, $H_{c2} = 1.12$ T and $k_{GL}=10.6$. The specific heat results demonstrate TISb to be a moderately coupled $s$-wave superconductor. Although we cannot determine which bands near $E_F$ are responsible for SC, the coexistence of bulk SC with $s$-wave symmetry and Dirac fermions on the surface in a single TISb compound provide an opportunity to realize the Majorana-zero energy mode.

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| Table 1 | Superconducting parameters of TISb |
|---------|---------------------------------|
| Parameters | Value |
| $T_C$ (K) | 4.3 |
| $H_{c1}(0)$ (Oe) | 148 |
| $H_{c2}$ (T) | 1.12 |
| $H_{c2}$ (Oe) (T) | 8.18 |
| $\xi_{an}$ (nm) | 15.3 |
| $\xi_{an}$ (nm) | 162 |
| $k_{GL}$ | 10.6 |
| $\gamma_0$ (mJ mol$^{-1}$ K$^{-2}$) | 5.56 |
| $\Delta C_{el}/\gamma_0 T_C$ | 1.42 |

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