Superconductivity in centrosymmetric topological superconductor candidate TaC

D Y Yan\textsuperscript{1,2}∗, M Yang\textsuperscript{1,2}∗, C X Wang\textsuperscript{1,2}, P B Song\textsuperscript{1,2}, C J Yi\textsuperscript{1,4} and Y G Shi\textsuperscript{1,3,4}

\textsuperscript{1} Beijing National Laboratory for Condensed Matter Physics and Institute of Physics, Chinese Academy of Sciences, Beijing 100190, People’s Republic of China
\textsuperscript{2} School of Physical Sciences, University of Chinese Academy of Sciences, Beijing 100190, People’s Republic of China
\textsuperscript{3} Center of Materials Science and Optoelectronics Engineering, University of Chinese Academy of Sciences, Beijing 100049, People’s Republic of China

E-mail: ygshi@iphy.ac.cn and chyi@iphy.ac.cn

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Abstract

We report the synthesis and physical properties of the single crystals of TaC, which are proposed to hold topological band structure as a topological superconductor (TSC) candidate. Magnetization, resistivity and specific heat measurements are performed and indicate that TaC is bulk superconductor with critical temperature of 10.3 K. TaC is a strongly coupled type-II superconductor and the superconducting state can be well described by \(s\)-wave Bardeen–Cooper–Schrieffer theory with a single gap. The upper critical field \((H_{c2})\) of TaC shows linear temperature dependence, which is quite different from most conventional superconductors and isostructural NbC, which is proposed to manifest topological nodal-loops or type-II Dirac points as well as superconductivity. Our results suggest that TaC would be a new candidate for further research of TSCs.

Keywords: \(s\)-wave BCS, topological superconductor, single crystal

(Some figures may appear in colour only in the online journal)

1. Introduction

Exploration of topological superconductors (TSCs) is progressively becoming a fruitful subject in condensed matter physics. A TSC has topologically nontrivial superconducting phases, characterized by a full superconducting gap in the bulk, but supporting a protected Majorana zero-energy mode (MZM) at the vortex core, as well as gapless edge or surface states [1–3]. Because the Majorana bound state obeys non-Abelian statistics and is theoretically predicted to emerge in certain \(p\)-wave TSCs, it provides a possible way to approach fault-tolerant quantum computation [4]. The topological surface state (TSS) and MZM have been found to exist in single material platforms of FeTe\textsubscript{0.55}Se\textsubscript{0.45} bulk single crystals [5–8] and similar compounds of iron-based superconductors [9–11]. In a TSC, it is the wave function of the electron pairs that exhibits topological properties, and among the most promising candidates for TSCs are materials with noncentrosymmetric (NCS) crystal structures. NCS crystal structure with broken inversion symmetry has antisymmetric spin–orbit coupling and spin degeneracy, which gives rise to topological phases and allowing the mixing of spin-triplet and spin-singlet pairing channels [12–14]. As yet, the effort to search for TSCs continues, but few topological NCS superconductors have been identified. In recent years, the observation of TSSs in some centrosymmetric superconducting materials has attracted most interest. For centrosymmetric superconductors, the parity mixing of pair potential may occur near the surface where the inversion symmetry

\(*\) To whom correspondence should be addressed.
is broken. Hence, a centrosymmetric superconductor may become a great candidate for TSC [15]. It has been found that the centrosymmetric $\beta$-PbBi$_2$ has TSSs at $E_F$ in the normal state and fully gapped superconductivity in the bulk, which makes it a great candidate for achieving TSC states [16–21]. Meanwhile, the anisotropic Majorana bound states were theoretically predicted and experimentally observed in 2M-WS$_2$, which is a centrosymmetric superconductor with $T_c \sim 8.8$ K [22, 23]. Si$_3$TaS$_2$ has also been identified to be a superconductor with a possible topological nodal line semimetal character [24]. Noteworthy, Cui et al proposed that type-II Dirac semimetal states exist in the band structure of TaC [25]. Shiroka et al reported the superconducting properties of the polycrystalline NbC and TaC and calculated their topological band structures, pointing out the potential TCSs states among these compounds [26]. Meanwhile, combining the theoretical calculation and angle resolved photoemission spectroscopy (ARPES) measurements, we observe that NbC is a type-II Dirac semimetal with robust Fermi-surface nesting that is responsible to the strong electron–phonon interaction [27].

In this work, we successfully grow the single crystals of TaC and report the magnetism, resistivity and specific heat properties. The results indicate that TaC is Bardeen–Cooper–Schrieffer (BCS) superconductor with s-wave single gap in bulk state. However, the electron–phonon coupling (EPC) strength of TaC is larger than most conventional BCS superconductors. In addition, the upper critical field shows anomalous linear temperature dependence at low temperature, which is usually present in some iron-based superconductors.

2. Experiment details

Single crystals of TaC were grown by using Co-flux method. Starting materials of high-purity Ta, C and Co were mixed and loaded in an alumina crucible at a molar radio of TaC:Co = 1:1:9. The alumina crucible was then placed in an argon-filled furnace and heated to 1500°C. After a dwell time of 20 h, the crucible was slowly cooled to 1300 °C at a rate of 1 °C h$^{-1}$ and then cooled naturally down to room temperature. Finally, the excess Co was removed by immersion in nitrohydrochloric acid for one day, yielding golden TaC single crystals in rectangle shape.

To investigate the crystalline structure, single-crystal x-ray diffraction (XRD) was carried out on Bruker D8 Venture diffractometer at 293 K using Mo $K\alpha$ radiation ($\lambda = 0.71073$ Å). The crystalline structure was refined by full-matrix least-squares method on $F^2$ by using the SHELXL-2016/6 program. Selected crystals were used for magnetic susceptibility ($\chi$), electrical resistivity ($\rho$) and specific heat ($C_P$) measurements. The magnetic properties were measured in a Magnetic Properties Measurement System (MPMS-III, Quantum Design Inc.) under fixed applied magnetic field of 20 Oe in field-cooling (FC) and zero-FC modes. Isothermal magnetization ($M$-$H$) was measured at several fixed temperatures by sweeping applied field. The electrical resistivity and specific heat were measured in a physical property measurement system (Quantum Design Inc.) by using a standard dc four-probe technique and a thermal relaxation method, respectively.

3. Result and discussion

The single-crystal XRD study reveals that TaC crystallize in a centrosymmetric NaCl-type structure with space group of $Fm\bar{3}m$ (No. 225). The detailed crystallographic parameters are summarized in table 1. Figure 1(a) shows the photograph of TaC crystal, and the back square of $1 \times 1$ mm indicates the size of the crystal. A schematic drawing of the crystal structure is shown in figure 1(b). The XRD patterns of a flat surface of the single crystal is presented in figure 1(c), where only the $h00$ peaks are detected. And the Laue diffraction pattern with fourfold symmetry on the (100) surface of TaC is presented in the inset of figure 1(c), further proving the quality of the single crystal.

Temperature dependent magnetic susceptibility ($4\pi \chi$) with an applied field $H = 20$ Oe was measured from 2 K to 16 K, as shown in figure 2(a). The critical temperature $T_c$ was determined by the intersection between the normal state of $4\pi \chi$ extrapolated to lower temperature and the superconducting state corresponding to the steepest slope of the diamagnetic area [28], yielding that $T_c$ 10.3 K for TaC. The superconducting volume fraction calculated from the original magnetic measurement exceeds 150%, which comes from the demagnetization field due to the sample. Using the following expression [29]:

$$\chi = \frac{\chi_{exp}}{1 - N\chi_{exp}},$$

where $N = 0.27$ is the demagnetizing factor estimated for the cuboid sample [30], the modified superconducting volume fraction is closed to 100%, as shown in figure 2(a). Figure 2(b) shows the $M$-$H$ loops in various temperatures for TaC, indicating that TaC is type-II superconductors [31].

The temperature dependence of resistivity measured from 300 K to 2 K is shown in figure 3(a) for TaC. The resistivity undergoes a sudden reduction near $T_c$. The onset and zero-resistance temperature were marked in figure 3(a). The zero-resistance temperature is consistent with the $T_c$ measured from magnetic susceptibility. Above $T_c$, the resistivity metallically goes down with cooling temperature, and the data can be well described by Bloch–Grüneisen law [32–35] with a formula of:

$$\rho(T) = \rho_0 + \alpha \left(\frac{T}{T_D}\right)^5 \int_0^{\theta_D/T} \frac{x^5}{(e^x - 1)(1 - e^{-x})} dx$$

### Table 1. Atomic coordinates and equivalent isotropic thermal parameters of TaC.

| Site | Wyckoff | x | y | z | Occupa | $U_{eq}$b |
|------|---------|---|---|---|--------|----------|
| Ta   | 4a      | 0 | 0 | 0 | 1      | 0.017(3) |
| C    | 4b      | 0 | 0.5 | 0 | 1      | 0.03(2)  |

*a Occupancy.

*b $U_{eq}$: equivalent isotropic thermal parameter.
Figure 1. (a) Photograph of a typical TaC single crystal. (b) The schematic crystalline structure of TaC. (c) The XRD patterns of a flat surface of TaC single crystals. The inset shows the Laue diffraction pattern on the (100) surface.

where $\rho_0$ is the residual resistivity of the normal state, $\alpha$ is proportional to electron–phonon coupling constant and $\theta_D$ is Debye temperature. The fit is shown as a red solid line in figure 3(a), giving out the Debye temperature $\theta_D = 213.5$ K. The figure 3(b) shows the magnetic field dependent resistivity at various fixed applied magnetic fields, indicating that the superconductivity is suppressed when magnetic field is applied. It should be noticed that the electrical resistivity shows multiple transitions by increasing the magnetic field. The second transition may be caused by the carbon-deficient in TaC single crystal, as previously reported that the superconducting transition temperature decreases with decreasing C content in TaC$_x$ ($0 < x < 1$) [36, 37].

With fitted parameters of $\theta_D$ and $T_c$, the EPC constant $\lambda_{ep}$ can then be calculated by using the inverted McMillan equation [38]:

$$\lambda_{ep} = \frac{1.04 + \mu^* \ln \left( \frac{\rho_0}{1.45T_c} \right)}{(1 - 0.62\mu^*) \ln \left( \frac{\rho_0}{T_c^{3.5}} \right) - 1.04}$$

where the $\mu^*$ represents the repulsive screened Coulomb part, which is set to 0.13 for transition metal element. This yields a value of the superconducting parameter $\lambda_{ep} = 0.986$. Typically, materials with $\lambda_{ep} \rightarrow 1$ are classified as strongly coupled superconductors, while $\lambda_{ep} \rightarrow 0.5$ indicates weak coupling [39]. The relatively large $\lambda_{ep}$ indicates strong EPC strength in TaC. The large EPC constant was also observed in the isostructural compounds NbC [27] and NbC$_{1-x}$N$_x$ [40], which may be caused by the existence of Fermi-surface nesting at the Fermi level, leading to the emergence of Kohn anomaly and enhance the EPC.

The specific heat data in various applied fields are given in figure 4(a) for TaC. With increasing fields, the superconducting peak is gradually suppressed to lower temperature, which is consist with the behavior in resistivity as aforementioned. Figure 4(b) plots $\Delta C_P = C(0T) - C(0.5T)$ versus $T$. Under the applied magnetic field, the jump of $C_P$ due to superconducting transition is totally suppressed. Since phonon and normal state electronic specific heat are field independent, $\Delta C_P$ is the specific heat after subtracting the background originating from the lattice and normal state
is the quasiparticle contribution and state Sommerfeld coefficient of the superconducting part. As seen in figure 4(b), the s-wave model well reproduces the experimental data, while the d-wave model deviates significantly from the data at low temperature. This result is same as NbC [27]. From the s-wave fit results, we get the parameters \( r = 1.17, \gamma_n = 2.21 \text{ mJ mol}^{-1} \text{K}^{-2} \) and \( T_c = 9.7 \text{ K} \) for TaC. It should be note that the \( T_c \) of 9.7 K gotten from specific heat data is smaller than the data 10.3 K measured from magnetic and resistivity data for TaC. Consequently, the estimated superconducting gap \( \Delta(0) \) is 1.72 meV. The entropy-conserving construction at \( T_c \) gives \( \Delta C/\gamma_n T_c = 1.94 \), which obviously deviates from the weak coupling BCS value 1.43. And the value of \( 2\Delta(0)/k_BT_c \) is 4.12, which is also slightly larger than the BCS theory value 3.52.

The phase diagram of the upper critical field \( H_{c2}(T) \) and the lower critical field \( H_{c1}(T) \) versus \( T \) are plotted, as shown in figure 5(a). \( H_{c1}(T) \) was defined from the point deviating from the \( M-H \) linear curve due to the Meissner effect. As shown in the inset of figure 5(a), the \( H_{c1}(T) \) were fitted by the empirical power law expression \( \mu_0H_{c1}(T) = \mu_0H_{c1}(0) \left[ 1 - \left( \frac{T}{T_c} \right)^b \right] \) with fixed \( T_c = 10.3 \text{ K} \). The fits reveal that \( \mu_0H_{c1}(0) = 38.6 \text{ mT} \). The upper critical field \( H_{c2} \) was defined from the 90% and 50% of the normal state resistivity as well as the zero point of the resistivity drop in figure 3(b) for every magnetic field. Furthermore, the upper critical field was determined from the \( M-H \)

**Figure 2.** (a) Temperature dependence of \( 4\pi \chi \) for applied field \( H = 20 \text{ Oe} \) in ZFC and FC modes. (b) Isothermal \( M-H \) curves of TaC at various fixed temperatures.

**Figure 3.** (a) Temperature dependence of longitudinal resistivity \( \rho \) of TaC. (b) The resistivity at various magnetic fields.
curves in figure 2(b) and the specific heat data in figure 4(a). One can see that the \( H_{c2} \) data of 90\%\( \rho \) and 50\%\( \rho \) very strangely deviate from other data, which needs further detailed studies for explanation. The \( H_{c2}(T) \) curve of TaC determined from zero resistivity, magnetization and specific heat data shows an almost linear \( T \)-dependence at low temperature, which cannot be fitted by WHH model \cite{44,45} and NbC single crystals reported by us \cite{27}, as shown in figure 5(b). The linear \( T \)-dependence of \( H_{c2}(T) \) is similar to some iron-based superconductors with multiband superconductivity \cite{46,47}. Considering the second transition in the resistivity measurement under magnetic field, the linear \( H_{c2}(T) \) may also be caused by the existence of the carbon-deficient in TaC. By linear extrapolations, the upper critical field is estimated to be 0.3 T, which is obviously smaller than the Pauli paramagnetic limit of 19 T, indicating that the orbital pair breaking is the essential mechanism and limits the upper critical field in TaC.

In addition, other parameters related to superconductivity can be calculated. By using the density of electronic states at the Fermi energy \( N(E_F) \) can be calculated from the formula \cite{48}:

\[
N(E_F) = \frac{3\gamma_{\text{a}}}{\pi^2\hbar^2} (1 + \lambda_{\text{ep}}). 
\] (5)

Thus the \( N(E_F) \) was estimated to be 0.69 eV\(^{-1}\) per formula unit (f.u.). The mean free path \( l \) can be determined by using following equation \cite{49}:

\[
l = 2.372 \times 10^{-14} \left( \frac{m^*}{m} \right)^2 \frac{V_M^2}{N(E_F)^2 \rho_0} \] (6)

where \( V_M \) is the molar volume. Assuming that \( m^*/m = 1 \), the \( l \) was calculated to be 448 Å.

The determined \( H_{c2}(0) \) value can be used to calculate the Ginzburg–Landau coherence length \( \xi_{\text{GL}} \) by using the equation \cite{31}:

\[
\mu_0 H_{c2}(0) = \frac{\Phi_0}{2\pi\xi_{\text{GL}}^2}. 
\] (7)

where \( \Phi_0 \) is the quantum flux \( \frac{h}{2e} \). The value of \( \xi_{\text{GL}} \) for TaC was calculated to be 331 Å.

Using the result of \( \xi_{\text{GL}} \) with \( H_{c2}(0) \), the Ginzburg–Landau penetration depth \( \lambda_{\text{GL}} = 595 \) Å of TaC were estimated by the lower critical field equation \cite{50}:

\[
\mu_0 H_{c1}(0) = \frac{\Phi_0}{4\pi\lambda_{\text{GL}}^2} \ln \left( \frac{\lambda_{\text{GL}}}{\xi_{\text{GL}}} + 0.49693 \right). 
\] (8)

From the Ginzburg–Landau penetration depth and the coherence length, the Ginzburg–Landau parameter is \( \kappa_{\text{GL}} = \frac{\lambda_{\text{GL}}}{\xi_{\text{GL}}} > \frac{1}{\sqrt{2}} \), which again confirms that TaC is a type-II superconductor \cite{31}. Combining the results of \( H_{c1}(0) \), \( H_{c2}(0) \) and \( \kappa_{\text{GL}} \), the thermodynamic critical field \( H_{c1}(0) \) was estimated to be 136 mT from the equation \cite{31}:

\[
H_{c1}H_{c2} = \frac{\mu_0 H_{c2}}{\xi_{\text{GL}} + 0.08}. 
\] (9)

The relationship between the BCS coherence length \( \xi_0 \) and the Ginzburg–Landau coherence \( \xi_{\text{GL}} \) at \( T = 0 \) K is \[51\]

\[
\frac{\xi_{\text{GL}}(0)}{\xi_0} = \frac{\pi}{2\sqrt{3}} \left( 1 + \frac{\xi_0}{\xi_{\text{GL}}} \right)^{-0.5}. 
\] (10)

From the above equation, we get the value of \( \xi_0 \) is 543 Å. The value of \( \xi_0/l \) is close to 1, indicating that TaC superconductor is on the boundary between the dirty and clean limits. In summary, the observed and estimated superconducting parameters of TaC and NbC are listed in table 2 for comparison.

Superconductors with nontrivial band structure provide possibility of realizing TSCs. Superconductivity can be combined with novel band structures by charge carrier doping or heterostructures constructed by superconductors and topological insulators. For instance, topological insulator \( \text{Bi}_2\text{Se}_3 \) becomes superconductor when intercalated by Cu, Sr, or Nb atoms \cite{52–54}. And the heterostructure of NbSe\(_2/\text{Bi}_2\text{Se}_3 \) is reported to hold TSCs at its interface \cite{55,56}. However, doping can induce disorder and inhomogeneity effect, which hinder the unambiguous clarification of superconducting states in doped topological insulators. The difficulty of fabricating heterostructure and observing the interface-related phenomena also limits further studies of TSC. Different from

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**Figure 4.** (a) Special heat \( C_p/T \) versus \( T \) curves at various applied magnetic fields. (b) Experiment data for \( \Delta C_p \) vs \( T \), plotted by fits using \( s \)-wave and \( d \)-wave gap function. The inset shows an enlarged image at low temperature.
Figure 5. (a) Phase diagram of the upper critical field \(H_{c2}\) of TaC versus \(T\) with the fitting curves. The inset shows the lower critical field \(H_{c1}\) versus \(T\). (b) Phase diagram of NbC from [27].

Table 2. Measured and calculated superconducting parameters of TaC and NbC.

| Parameter               | TaC   | NbC [27] |
|-------------------------|-------|----------|
| \(T_c\)                | 10.3 K| 11.5 K   |
| \(\mu_0H_{c1}(0)\)     | 38.6 mT| 19.6 mT  |
| \(\mu_0H_{c2}(0)\)     | 0.3 T | 1.21 T   |
| \(\mu_0H_{c3}(0)\)     | 136 mT| 107 mT   |
| \(l\)                   | 448 Å | 33 Å     |
| \(\xi_0\)               | 543 Å | 1035 Å   |
| \(\xi_{GL}\)           | 331 Å | 165 Å    |
| \(\lambda_{GL}\)       | 595 Å | 1322 Å   |
| \(\kappa_{GL}\)        | 1.8   | 8.01     |
| \(\gamma_n\)           | 2.21 mJ mol\(^{-1}\)K\(^{-2}\) | 3.01 mJ mol\(^{-1}\)K\(^{-2}\) |
| \(\Delta C/\gamma_nT_c\)| 1.94  | 1.43     |
| \(\mu_0H_p\)           | 19 T  | 21 T     |
| \(\lambda_p\)          | 0.986 | 0.848    |
| \(N(E_F)\)             | 0.47 eV\(^{-1}\) per f.u. | 0.69 eV\(^{-1}\) per f.u. |
| \(\theta_D\)           | 213.5 K| 321.6 K |
| \(r\)                  | 1.17  | 1.06     |
| \(\Delta(0)\)          | 1.72 meV | 1.85 meV |
| \(2\Delta(0)/k_BT_c\)  | 4.12  | 3.73     |

the two strategies mentioned above, the coexistence of the bulk superconductivity and topological band structures in the same compound might generate intrinsic TSC with nontrivial surface states. For example, nontrivial topological band structures have been observed in \(\beta\)-PdBi\(_2\) [16] and PbTaSe\(_2\) [15] superconductors, which provide excellent playground for studying TSC. However, their superconducting temperature are relatively low (<5 K), limiting the deeper investigations and possible applications.

Our experiments prove that TaC is a BCS superconductor with single \(s\)-wave gap in bulk. It has simple structure and high resistance to corrosion with high \(T_c\) of 10.3 K. Meanwhile, TaC shows some unusual superconductivities such as strong EPC constant and linear temperature dependence of \(H_{c2}\), which is distinct from most conventional superconductors as well as the isostructural NbC single crystal we reported recently [27]. The theoretical studies and ARPES measurements for NbC confirm that the superconductivity as well as the topological band structures could emerge together in this compound. The Fermi-surface nesting was observed in NbC by APPES, which is corresponding to the strong EPC. However, the ARPES research for TaC is still absent due to the difficulties on cleavage. First-principles calculation of TaC by Cui et al indicates the type-II Dirac point near \(\Gamma\) point in the Brillouin zone [25]. They also claim that Ta \(d\) orbitals and C \(p\) orbitals consist the gapless Dirac point with nontrivial topology. Therefore, the successful synthesis of single crystals of TaC may enlarge the measureable materials family of TSCs and provide new opportunity for further research on novel physical properties.

4. Conclusion

In summary, we present the studies of magnetism, resistivity and specific heat on single crystal TaC, which is characterized as a type-II superconductor with centrosymmetric structure. The electronic specific heat in the superconducting state is well described by the single gap \(s\)-wave BCS expression. Most of superconducting properties for TaC are similar to NbC single crystals we reported in [27]. The value of electron–phonon coupling constant is quite larger compared with conventional BCS superconductors, which may be caused by the Fermi-surface nesting in its electronic structures. Meanwhile, the linear behavior of \(H_{c2}\) vs \(T\) for TaC is different from NbC and most BCS superconductors, making TaC a valuable material for further study. Hence, we suggest that TaC is a potential candidate of TSCs for further investigating the novel physical properties.

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ORCID ids

D Y Yan https://orcid.org/0000-0001-6484-9968
M Yang https://orcid.org/0000-0003-2862-2015
C J Yi https://orcid.org/0000-0002-2659-2628

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