Electronic band tuning under pressure in MoTe$_2$ topological semimetal

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Topological superconductors (TSC) can host exotic quasiparticles such as Majorana fermions, poised as the fundamental qubits for quantum computers. TSCs are predicted to form a superconducting gap in the bulk, and gapless surface/edges states which can lead to the emergence of Majorana zero energy modes. A candidate TSC is the layered dichalcogenide MoTe$_2$, a type-II Weyl (semi) metal in the non-centrosymmetric orthorhombic ($T_d$) phase. It becomes superconducting upon cooling below 0.25 K, while under pressure, superconductivity extends well beyond the structural boundary between the orthorhombic and monoclinic ($1T'$) phases. Here, we show that under pressure, coupled with the electronic band transition across the $T_d$ to $1T'$ phase boundary, evidence for a new phase, we call $T_d'$ is observed and appears as the volume fraction of the $T_d$ phase decreases in the coexistence region. $T_d'$ is most likely centrosymmetric. In the region of space where $T_d'$ appears, Weyl nodes are destroyed. $T_d'$ disappears upon entering the monoclinic phase as a function of temperature or on approaching the suppression of the orthorhombic phase under pressure above 1 GPa. Our calculations in the orthorhombic phase under pressure show significant band tilting around the Weyl nodes that most likely changes the spin-orbital texture of the electron and hole pockets near the Fermi surface under pressure that may be linked to the observed suppression of magnetoconductivity with pressure.

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INTRODUCTION

A Weyl (semi)metal is a new topological state of matter that hosts the condensed matter equivalent of relativistic Weyl fermions. Weyl fermions exist as low-energy electronic excitations at Weyl nodes in three-dimensional momentum space, producing exotic physical properties such as unique surface Fermi arcs and negative magnetoresistance.$^{1-4}$ The topological Weyl state can be realized by breaking either time-reversal or lattice inversion symmetry. A candidate topological Weyl semimetal is the quasi two-dimensional transition metal dichalcogenide MoTe$_2$. The transition to the non-trivial topologically protected crystal state occurs upon cooling from the high temperature $1T'$ monoclinic phase to the low temperature orthorhombic $T_d$ phase. The transition is driven by $c$-axis layer stacking order around 250 K.$^{9-13}$ Upon cooling to the non-centrosymmetric $T_d$ phase, Weyl quasiparticles are expected at characteristic electron and hole band crossings in momentum space.

MoTe$_2$ is a candidate TSC in the orthorhombic phase at ambient pressure.$^{14}$ In the superconducting state, Fermi arcs are proposed to still exist even though the Weyl nodes are completely gapped out by the superconducting gap, and the onset of superconductivity can generate a quantum anomaly analogous to the parity anomaly in quantum electrodynamics.$^{15,16}$ The application of pressure enhances the superconducting transition temperature from 0.25 to about 8 K$^{15}$ and extends the superconducting phase over a wide pressure range. Recent studies showed that while in the superconducting state, a phase transition occurs from the orthorhombic $T_d$ to the monoclinic $1T'$ phase under pressure$^{17}$ with a large hysteresis region and coexistence of the two phases. A high-pressure muon-spin rotation study of superconductivity in MoTe$_2$ suggested that a topologically non-trivial $s^-$ state likely exists in MoTe$_2$ at pressures up to 1.9 GPa.$^{18}$ Key components to a TSC state are the crystal structure and band topology under pressure. Existing reports stop short of exploring the symmetry in the superconducting phase under pressure and no calculations exist on the electronic band structure. Using single crystal neutron diffraction, the pressure–temperature phase diagram is mapped out, and combined with band structure calculations, we elucidate the effects of pressure on the electronic band structure topology.

The results provide evidence for a topological state that persists under pressure, consistent with the muon work of ref. 18. MoTe$_2$ can crystallize into several different structures including the hexagonal $2H$, the monoclinic $1T'$ and the low temperature orthorhombic $T_d$ phases.$^{9-11}$ The hexagonal $2H$ phase (with space group $P6_3/mmc$), a semiconductor with an indirect band gap of 0.88 eV, is stable below 900 °C.$^{19,20}$ The monoclinic $1T'$ phase is stable above 900 °C but can be stabilized to room temperature by quenching.$^{9,10,20}$ In the $1T'$ phase, Mo is surrounded by Te in an octahedral environment with Mo shifted off-center. Mo atoms form zigzag chains running along the $b$-axis that distort the Te sheets resulting in a tilted $c$-axis with angle $\beta \approx 93^\circ$ (Fig. 1a).$^9$ Upon cooling from the $1T'$ and not from the $2H$ phase, a first order structural transition occurs into the orthorhombic $T_d$ phase (Fig. 1b). This transition comes about as a result of layer stacking order along the $c$-axis in the $T_d$ phase.$^{13}$

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Fig. 1  Physical properties. a Crystal structure of MoTe$_2$ in the 1T’ phase projected on the $ab$-plane, with the zigzag chains marked running along the $a$-axis. b Unit cell of the T$_d$ structure projected on the $ac$ plane. c Temperature-dependent superconducting (SC) shielding [zero-field-cooled (ZFC)] and Meissner (FC) fraction data for MoTe$_2$ in $H = 20$ Oe at 1.2 GPa. d Magnetic hysteresis loop for MoTe$_2$ at 2 K at 1.2 GPa. Arrows indicate sweeping direction. e, f Contour map of the neutron scattering intensity obtained by scanning across the (2 0 l) Bragg peak along the [0 0 1] direction at 0.15 GPa on cooling (e) and warming (f). The peak labels denoted with D1 (main domain) and D2 (second domain) are used to distinguish the peaks coming from two different domains, and subscripts 1T’ and T$_d$ are used to distinguish the peaks in the different phases. g Temperature dependence of the $\mathbf{q}$ integrated neutron scattering intensity for $(201)_{1T'}^0$ peak (red squares), $(201)_{1T'}^1$ peak (blue circles), and $(201)_{TD}^2$ (green circles). Arrows indicate sweeping directions. h, i Contour map of the neutron scattering intensity obtained by scanning across the (2 0 l) Bragg peak along the [0 0 1] direction at 0.95 GPa on cooling (h) and at 1.2 GPa on cooling (i). j The phase diagram under pressure. The region between the two phases shows coexistence of both phases plus the T$_{d}^{*}$ phase. Inversion symmetry is broken only in the T$_d$ phase while both T$_{d}^{*}$ and 1T’ are centrosymmetric. k A two-dimensional scan in the h0l plane outside the pressure cell with half-integer peaks from the T$_{d}^{*}$ phase appearing along L. The dashed white lines mark the integer peaks along L that correspond to both the T$_d$ and T$_{d}^{*}$ phases while the half-integer peaks belong to the T$_{d}^{*}$ phase only.
RESULTS

Shown in Fig. 1c, d are the plots of the bulk magnetic susceptibility, $\chi(T)$, and magnetization, $M$, under pressure as a function of temperature and magnetic field, respectively. Our crystal becomes diamagnetic below 3.0 K at 1.2 GPa, marking the onset of superconductivity. Bulk superconductivity is evident in the full shielding diamagnetic signal at 2.0 K. Note that $\chi(T)$ was measured along the $ab$-plane in which the demagnetizing factor is negligible. The crystal exhibits a large shielding fraction, $\sim100\%$ (zero-field-cooled (ZFC) magnetization), at 2 K and 1.2 GPa. Magnetic hysteresis loops were also measured at 2.0 K and 1.2 GPa as shown in Fig. 1d indicating that MoTe$_2$ exhibits a typical type-II superconducting behavior.

The 1T$'$ to Td phase transition under pressure was explored by single crystal neutron diffraction performed in the 20L scattering plane, focusing on structural signatures most affected by variations in the angle $\beta$ between the $a$- and $c$-axes. The crystal was aligned using the lattice constants and $\beta$ of the 1T$'$ symmetry ($a = 6.3363 \AA$, $b = 3.469 \AA$, $c = 13.7789 \AA$, $a = 90^\circ$, $\beta = 93.75^\circ$, $\gamma = 90^\circ$). A typical contour map of the scattering intensity as a function of temperature at constant pressure is shown in Fig. 1e at 0.15 GPa. At 300 K in the 1T$'$ phase, a very strong Bragg peak at $(hkl) = (2 0 1)$ is observed along with two weak secondary peaks, at $(0 4 0 1)$ and $(1 4 0 1)$. The Bragg peaks are from the two possible domains in the monoclinic symmetry and are marked with superscripts D1 and D2 indicating domain one and two, respectively. One domain (D1) is overwhelmingly more intense than the other and the crystal was aligned using the primary domain. At 0.15 GPa, the 1T$'$ to Td phase transition is clearly observed upon cooling, with a discontinuous volume change and the transition temperature shifting to a lower value compared to the ambient pressure value ($\sim$240 K). At the transition from 1T$'$ to Td at 220 K, the 1T$'$ peaks disappear and the Td peaks, $(201)_D$, $(201)_D$, and $(202)_D$ peaks disappear and the Td peaks, $(201)_D$ and $(202)_D$ appear as shown in Fig. 1e. The $(201)_D$ and $(202)_D$ peaks appear at different $\beta$ positions corresponding to the change of $\beta$ from 93.75$^\circ$ (1T$'$) to 90$^\circ$ (Td). There is only a single domain and one set of peaks in the Td phase. Alignment of the single crystal remained the same as at 300 K during cooling. On warming, a similar set of Bragg peaks is observed and shown in the contour map of Fig. 1f as on cooling but with an additional Bragg peak that arises from the new Td* phase. While only one peak at $(203)_D$ is present in this narrow range of the 20 L scan inside the pressure cell, other Td* peaks have been observed along L. This is shown in the 2D plot of the HOL scattering plane from a single crystal measurement at 300 K outside the pressure cell (Fig. 1k). Td* peaks appear at half-integer L-points and are prominent along 20 L and 30 L. No Td* peaks are visible on cooling which is consistent with earlier single crystal neutron diffraction measurements (see ref. 21). A thermal hysteresis is evident with the transition temperature shifting to 250 K on warming, which is higher than that on cooling, consistent with the thermal hysteresis observed at ambient pressure and in transport measurements. The temperature dependence of the integrated intensities for $(201)_D$, $(201)_D$ and $(201)_D$ Bragg peaks are shown in Fig. 1g, where coexistence of the two phases appears in the hysteresis region while the Td* phase only exists in the coexistence region. The structure parameters for the Td* phase obtained from the data outside the pressure cell are provided in Table 1. The Td* phase grows at the expense of the Td phase. At no point in pressure or temperature do we observed the Td* phase becoming the only phase, indicating that Td and Td* coexist. Td* disappears upon further warming into the 1T$'$ phase. At 0.15 GPa and 230 K data, all three phases coexist (Fig. 1g).

As the pressure increases to higher values, a precipitous decrease of the 1T$'$ to Td transition temperature occurs, eventually leading to the disappearance of the Td Bragg peaks by about 1.2 GPa. The intensity plot of Fig. 1h at 0.95 GPa clearly shows the phase transition temperature going down until its complete disappearance by 1.2 GPa as shown in Fig. 1i. At 1.2 GPa, only 1T$'$ Bragg peaks are observed in the temperature range from 5 to 300 K, regardless of the thermal cycling. Note that the two-domain feature of the 1T$'$ phase is observed at all pressure points. The single crystal results were additionally confirmed by neutron powder diffraction measurements under pressure that reached down to 1.5 K. The phase diagram under pressure is shown in Fig. 1j. Shown on the diagram are three distinct regions: the high temperature monoclinic region, the low temperature orthorhombic region and the coexistence region between the two that is also host to the new Td* phase. On warming, the Td* phase appears up to about 1 GPa; unit cell doubling occurs in the coexistence region of the two phases that disappears upon applying pressure greater than 1 GPa. This is shaded in green on the phase diagram. Above 1 GPa, a broad transition from the Td to 1T$'$ with apparent phase coexistence is observed. Coupled with the transition to the monoclinic phase under pressure is suppression of the magnetoresistance$^{22}$ which may be linked to the disappearance of the Weyl nodes.

Shown in Fig. 2 is a comparison of the Fermi surface projected on the $k_x$–$k_y$ momentum plane in the orthorhombic Td phase, calculated at two pressure points, 0 GPa (upper panels) and 0.15 GPa (lower panels). At 0 GPa, the two Weyl points indicated by dots on the $k_x$–$k_y$ plane at $k_z = 0$ and at $E = E_F = 23$ and 52.5 meV appear at the intersection of electron and hole pockets as indicated in both figures and also in the band structure diagram on the right where the crossing points in the dispersion are marked by the letters A and B. These Weyl nodes are the same as those reported in ref.$^{21}$. Increasing the pressure to 0.15 GPa changes the c-axis lattice constant by about 0.5% (with negligible change in the a-axis lattice constant). At 0.15 GPa, new nodes appear at $E = E_F = 18$ and 58 meV above the Fermi level, on the same band as the nodes at $E = E_F = 23$ and 52.5 meV at 0 GPa, but at $k_z = 0$. At 0.15 GPa, the superconducting transition temperature increases as reported in ref.$^{14}$ while magnetoresistance goes down.$^{22}$ The same nodes could not be traced in the band dispersions from calculations performed at higher pressures. At pressure values exceeding 1.2 GPa, the 1T$'$ symmetry is centrosymmetric and no Weyl nodes are expected. Ambient pressure calculations confirmed that the 1T$'$ with the P2$_1$/$m$ space group remains topologically trivial.$^{13}$ Thus, with warming from the Td to Td* and then to the 1T$'$ phase, the type-II Weyl nodes are destroyed.$^{13}$

| Table 1. Crystal unit cell parameters |
|--------------------------------------|
| Mo1 | Mo2 | Mo3 | Mo4 | Te1 | Te2 | Te3 | Te4 | Te5 | Te6 | Te7 | Te8 |
|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| $x$  | 0.893 | 0.746 | 0.321 | 0.321 | 0.502 | 0.355 | 0.578 | 0.578 | 0.147 | 0.000 | 0.932 | 0.932 |
| $z$  | 0.251 | 0.751 | 0.497 | 0.997 | 0.300 | 0.800 | 0.071 | 0.571 | 0.173 | 0.673 | 0.444 | 0.944 |

Atom coordinates of the proposed Td* structure. The space group is P21/m, with $a = 6.33 \AA$, $b = 3.47 \AA$, $c = 27.66 \AA$, and with the y-coordinates set at 0 for all atoms.
DISCUSSION

Analysis of the Berry curvature of the new nodes that appear in the Td phase from the band structure calculations performed at 0.15 GPa shown in Fig. 2 indicates that indeed these new crossings are not trivial. This is demonstrated in Fig. 3 which is a comparison of the Fermi surface cuts along k′ = 0 at 0.15 GPa in the Td phase and the two Weyl nodes in the Td phase at 0 and 0.15 GPa shown in Fig. 2. The Fermi surface morphology changes under pressure as seen from Fig. 2 while the Weyl nodes, observed closed to the Γ point and indicated by A and B, are not trivial. This is demonstrated in Fig. 3 which is a comparison of the Fermi surface cuts along k′ = 0 at 0.15 GPa in the Td phase and the two Weyl nodes in the Td phase at 0 and 0.15 GPa shown in Fig. 2. While the Weyl nodes observed closed to the Γ point and indicated by A and B are not trivial, they become significantly tilted under pressure, especially for the second Weyl node at 0.15 GPa shown in Fig. 2. The Fermi surface cuts shown on the left panels reveal the Weyl nodes at the intersections of electron and hole pockets. These are marked by A and B. The right panels are band structure plots showing the bands crossing at A and B.

METHODS

MoTe2 single crystals were grown by self-flux method. The details of single crystal growth have been described elsewhere. The typical size of the obtained single crystals is about 4 × 2 × 1 mm². Single crystal neutron diffraction under high pressure is combined with electronic band structure calculations to investigate the superconducting transition. Our MoTe2 single crystal exhibits a superconducting transition near 3 K at 1.2 GPa from bulk magnetic susceptibility measurements under pressure. The bulk magnetic susceptibility was measured at high pressure using a vibrating sample magnetometer (VSM) to demonstrate the superconducting transition in our MoTe2 single crystals. The applied pressure was calibrated by measuring pressure cell compression and the pressure dependence of the superconducting transition temperature of a tiny lead piece. High-pressure neutron diffraction experiments were performed on single crystals at HB-1A triple axis spectrometer and using powders at HB-2A powder diffractometer at the High Flux Isotope Reactor (HFIR) of Oak Ridge National Laboratory (ORNL). Self-clamped piston cylinder cells made with CuBe was used as pressure cells for both bulk magnetic susceptibility measurements and neutron diffraction experiments. Daphne 7373 oil was chosen as the pressure transmitting medium. Single crystal neutron
diffraction were performed at HB1 triple axis spectrometer with a fixed incident neutron energy of 13.5 meV. The lattice constant measurements of NaCl were used to calibrate the pressure. The single crystal neutron diffraction data shown in Fig. 1k were collected at the CORELLI spectrometer at ORNL.

Calculations
The theoretical DFT calculation was implemented in the Vienna Ab initio Simulation Package (VASP) for the pressurized Td and 1T’ phases of MoTe₂. VASP is a plane-wave based projector augmented pseudopotential local density functional method. Its exchange-correlation functional takes the generalized gradient approximation parameterized by Perdew-Burke-Ernzerhof. We turned on the spin–orbit coupling, used the default energy cutoff for the calculation and chose a K-grid of 13*7*3. An NBANDS of 160 is used to allow enough bands for Wannier90 to fit a minimum basis tight binding Hamiltonian involving Mo’s 4d and Te’s 5p orbits with necessary symmetrization. We used Wannier_tools for ab initio data processing, including nodal searching, Chirality calculation, and Fermi surface contour

![Graphs and data plots](image-url)
The neutron data collected for this project can be made available to interested researchers upon request. Similarly for the data collected for bulk characterization.

# Table 2. Electronic structure parameters

| P (GPa) | Td |
|---------|----|
|         |    |
| 0       | 0.0537 | 0.0053 | 0.058 | 0.0187 |
| 0.15    | 0.1071 | 0.1091 | 0.1007 | 0.101 |
| 0       | 0.0548 | 0.0227 | 0.0478 | -0.0135 |
| 0.15    | 0 | 0 | 0 | 0.0907 |
| 0       | 0.0045 | 0.0595 | 0.0340 | -0.2674 |
| 0.15    | 0.4754 | 0.3035 | 0.0655 | -0.2341 |
| 0       | 5.9155 | 6.1564 | 5.8641 | -4.4762 |
| 0.15    | -0.0045 | -0.0596 | -0.0340 | -0.3415 |
| 0       | -0.3575 | -0.3443 | -0.6858 | -0.6715 |
| 0.15    | 1.5610 | 3.0122 | 1.6813 | -7.0697 |
| 0       | 74.36 | 65.11 | 70.94 | -83.94 |
| 0.15    | -90.00 | -90.00 | -90.00 | -4.03 |
| 0       | -79.87 | -71.38 | -80.21 | -84.70 |

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# AUTHOR CONTRIBUTIONS

S.D. led the neutron experiment under pressure and contributed to the writing of the paper. M.M. contributed to the neutron experiment. J.Y. and J.A.S. contributed samples for this experiment. J.Y. led the characterization of superconductivity under pressure, and contributed to the writing of the paper. J.L. and C.Y. carried out the density functional theory calculations. C.D. and J.C.Y. Teo carried out the k.p calculations. C.D. and J.A.S. contributed to the identification of the Td phase. D.L. wrote the paper and led the project.

# ADDITIONAL INFORMATION

Competing interests: The authors declare no competing interests.

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