

Spin-Resolved Electronic Response to the Phase Transition in MoTe₂

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The semimetal MoTe₂ is studied by spin- and angle-resolved photoemission spectroscopy across the centrosymmetry-breaking structural transition temperature of the bulk. A three-dimensional spin-texture is observed in the bulk Fermi surface in the low temperature, noncentrosymmetric phase that is consistent with first-principles calculations. The spin texture and two types of surface Fermi arc are not completely suppressed above the bulk transition temperature. The lifetimes of quasiparticles forming the Fermi arcs depend on thermal history and lengthen considerably upon cooling toward the bulk structural transition. The results indicate that a new form of polar instability exists near the surface when the bulk is largely in a centrosymmetric phase.

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MoTe₂ exhibits a range of phenomena intersecting the physics of polar lattice transitions, topological phases of matter, and novel magnetoelectric properties. The centrosymmetric 1T' crystal undergoes a first-order transition into the noncentrosymmetric T_d structural phase upon cooling through T_S ≈ 250 K, with volume fractions of both phases appearing within the 200–300 K range [1–5]. Such transitions are very rare in metals and allow for control over the appearance of Weyl semimetal phases of matter (WSPs) and momentum dependent spin polarization (spin texture) that would be desirable for spintronic applications [6]. Superconductivity proposed to be topologically nontrivial has been observed [7–10]. Like WTe₂ [11,12], T_d-MoTe₂ is a type-II Weyl semimetal candidate [13,14] and exhibits extreme transverse magnetoresistance (XMR) with turn-on behavior [15–17]. Simultaneous tuning of electronic properties and the structural transition temperature and the breadth of the mixed-phase region is realized as a function of doping [3] and pressure or strain [5,9,18]. The sizes and shapes of the bulk electron Fermi pockets (EPs) and hole Fermi pockets (HPs) are important to the electronic basis for the properties of (Mo/W)Te₂ [19], but there is growing recognition that responses of electronic state vectors, described in terms of their spin and/or orbital projections, play a central role [16,18,20–22].

The WSP is predicted to be sensitive to the lattice parameters [13,14,23] and cannot exist in the centrosymmetric 1T' crystal, wherein all of the bulk bands must be spin-degenerate. However, the electronic structure of 1T'-MoTe₂ (T > T_S) observed in photoemission

spectroscopy appears much the same as that of T_d-MoTe₂ (T ≪ T_S) [24], although the decay of photoexcited states is clearly affected (likely due to loss of the WSP) [25]. Different reports on T_d-MoTe₂ favor the case of zero (trivial semimetal) [26], four [14,27–29], or eight [23,30–34] Weyl points (WPs) in the Brillouin zone (BZ) at locations ranging from approximately 5 [33] to 55 meV [28] above the Fermi energy E_F. The WPs impose subtle constraints on surface Fermi arc dispersions in (Mo/W)Te₂ systems [23,35–37], which have been taken as experimental signatures of the WSP [23,24,27,29–36,38–40]. Two types of Fermi arc are present [23,24,28]. Small arcs are buried within the HPs and a large arc appears in the gap between the HPs and EPs. The large arc persists in 1T'-MoTe₂ [24] and in the absence of WPs in WTe₂ [28,35], reinforcing the fact that Fermi arcs provide insufficient (although necessary) evidence of a WSP [41]. Quasiparticle scattering of the Fermi arcs is strongly affected by the structural transition [29]; however, this scattering occurs as a function of spin texture and nesting conditions rather than being directly related to the WSP [28].

Spin-texture visualization provides a resource for understanding scattering amplitudes, spin transport [42], MR anisotropy [20], and the pairing order and critical field enhancement in superconductivity [43]. Spin- and angle-resolved photoemission spectroscopy (SARPES) was used to probe the spin texture of T_d-(Mo/W)Te₂ in a few instances [24,30,34,44], but only small areas of momentum space were covered without measuring the full spin-polarization vector **P**. In this Letter, SARPES measurements and density functional theory (DFT) calculations

reveal a spin texture in the T_d -MoTe₂ Fermi surface that is three dimensional (3D) both in terms of spin orientation and momentum dependence. Small and large Fermi arc states persist at more than 90 K above T_S . Their coherence improves significantly upon cooling through T_S , and the appearance of the large Fermi arc state is affected by thermal history. Both an anomalous trend of Fermi arc self-energy through the transition and the residual spin polarization in bulk electrons at $T > T_S$ suggest that T_d and $1T'$ structural phases coexist near the surface at room temperature.

Details of the crystal synthesis and DFT calculations are provided in the Supplemental Material (SM) [45]. Experiments were performed with the sample kept under an ultrahigh vacuum (UHV) (pressure $< 1 \times 10^{-9}$ Pa) at variable temperatures fully summarized in the SM. Temperature was measured using a Si diode near the sample. Clean (001) surfaces were obtained by cleaving in UHV. High resolution spectra were obtained using a Scienta R4000 analyzer with instrumental angle and energy resolution better than 0.1° and 10 meV. SARPES measurements were done at the COPHEE endstation [67] with angle and energy resolution better than 1.5° and 75 meV. No evidence of mixed (001) and (00 $\bar{1}$) terminations [23,34] was found in our samples [68]. ARPES and quasiparticle interference results were consistent with only one termination type [28].

Figure 1 captures the DFT-calculated T_d -MoTe₂ Fermi surface and its spin texture, computed as an average over the orbital degree of freedom. The HPs enclosing the Γ point of the BZ and EPs located further from the Γ point

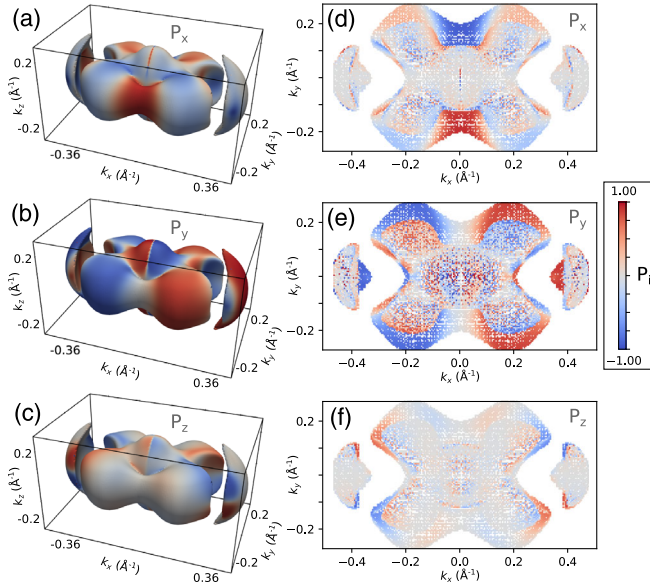


FIG. 1. Results of first-principles calculations for T_d -MoTe₂. False-color maps of (a) P_x , (b) P_y , and (c) P_z on the full bulk Fermi surface and the corresponding average of (d) P_x , (e) P_y , and (f) P_z over the interval $-\pi < k_z < 0$ projected into the k_x, k_y plane.

both exhibit high spin polarization reaching up to 0.8 in total magnitude [45]. This indicates significant orbital anisotropy when compared with, e.g., the Bi₂Se₃ surface state ($|\mathbf{P}| = 0.5$) [69]. The magnitudes of computed and measured spin-polarization have different significance, because photons selectively excite or entangle orbital components of the electron wave function [45,70,71], but the spin orientations will be shown to transform according to crystal symmetries in the same way for both cases. In addition to time-reversal symmetry, the space group contains one reflection M_x and one glide reflection M_y , which take the spatial coordinates (x, y, z) to $(-x, y, z)$ and $(x, -y, z + c/2)$, respectively, where c is the unit cell length perpendicular to the plane of the MoTe₂ layers. The in-plane components of \mathbf{P} are constrained by M_y and M_x such that $P_x \rightarrow -P_x$ as $k_y \rightarrow -k_y$ and $P_y \rightarrow -P_y$ as $k_x \rightarrow -k_x$, respectively. P_z reverses sign under both reflections, and also reverses under $(k_x, k_y, k_z) \rightarrow (k_x, k_y, -k_z)$. This is a novel property that suppresses $k_z \rightarrow -k_z$ scattering [45]. The spin polarization remains significant in all components when averaged over the lower half of the BZ as shown in Figs. 1(d)–1(f). This is important for SARPES because the k_z resolution is limited to about 35% of the reciprocal lattice vector due to the finite probing depth.

Figure 2(a) shows photon energy dependence of ARPES intensity at E_F along $\Gamma\bar{X}$. The states disperse with photon energy, thus characterizing their bulk (k_z dispersive) origin

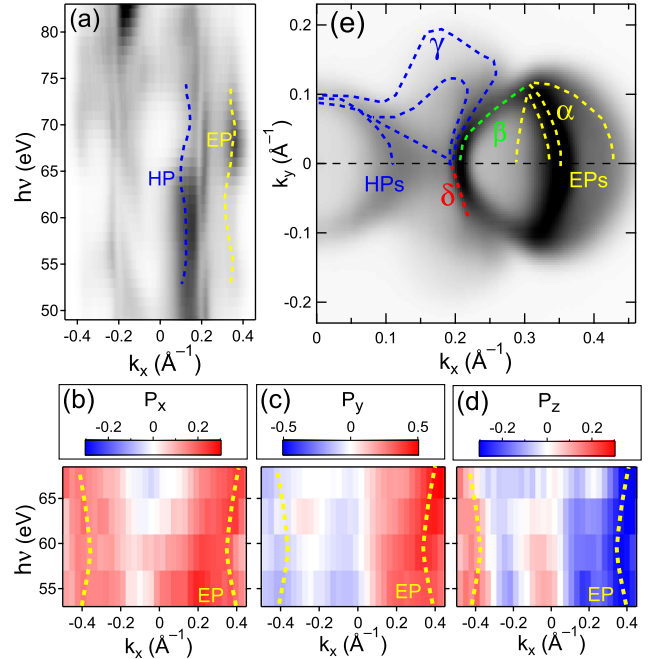


FIG. 2. Photoemission data collected for T_d -MoTe₂ at $T = 30$ K. Photon energy dependence at E_F of (a) ARPES intensity along $\Gamma\bar{X}$ (b)–(d) spin polarization measured at $k_y \approx 0.05 \text{ \AA}^{-1}$ for (b) P_x , (c) P_y , and (d) P_z . (e) Symmetrized ARPES intensity at E_F in the k_x, k_y plane.

[27]. The EP around $k_x = \pm 0.4 \text{ \AA}^{-1}$ (yellow dashed lines) produces the strongest spin-polarization signal seen in the photon energy dependent maps in Figs. 2(b)–2(d), which are taken along the same direction as in Fig. 2(a), but with a slight misalignment to $k_y \approx 0.05 \text{ \AA}^{-1}$. The signs of (P_x, P_y, P_z) for the EP are $(+, +, -)$ for positive k_x and $(+, -, +)$ for negative k_x , as enforced by the M_x symmetry. The experimental Fermi surface in the (k_x, k_y) plane is shown in Fig. 2(e) in which the contours of bulk EPs (yellow dashed lines), HPs (blue dashed lines), large Fermi arc (green dashed line), and small Fermi arc (red dashed line) are indicated. The states making up the largest EP, the large Fermi arc, the largest HP, and the small Fermi arc are labelled α , β , γ , and δ , respectively.

Figures 3(a)–3(d) show SARPES measurements of the Fermi surface along the k_y direction for $k_x = 0.26 \text{ \AA}^{-1}$, which crosses through β and γ . The contributions of these states to the spin polarization shown in Figs. 3(a)–3(c) and intensity in Fig. 3(d) were disentangled quantitatively by vectorial analysis [72] for the case of $T = 30 \text{ K}$. The momentum distribution curves (MDCs) were fit using four

Voigt peaks, two for β and two for γ , on a uniform unpolarized background and assuming $|\mathbf{P}| = 1$ in each peak. The inset in Fig. 3(d) shows the P_x , P_y , and P_z values obtained for each peak in green, orange, and purple bars, respectively. The P_x signal primarily originates from hole-like states in this momentum cut, as seen in the binding energy dependence of P_x in Fig. 3(l). The fit results show that β and γ have opposite signs of P_y . It is crucial to recognize that the sign of P_z reverses upon $k_y \rightarrow -k_y$ in both β and γ . This indicates that these states are constrained by bulk M_y symmetry, which is broken on the T_d -MoTe₂(001) surface [45]. The quality of the fit with $|\mathbf{P}| = 1$ in each state indicates fully coherent spin-orbital coupling at $T = 30 \text{ K}$.

Figures 3(e)–3(p) show measurements taken before and after cooling from 300 to 30 K. The spin polarization at E_F for the two temperatures is also compared in Figs. 3(a)–3(c). Response to the temperature change is evident in the lack of a coherent contribution from β and overall suppression of P_y and P_z at 300 K. The P_x signal of the hole-like states is retained through the full energy range

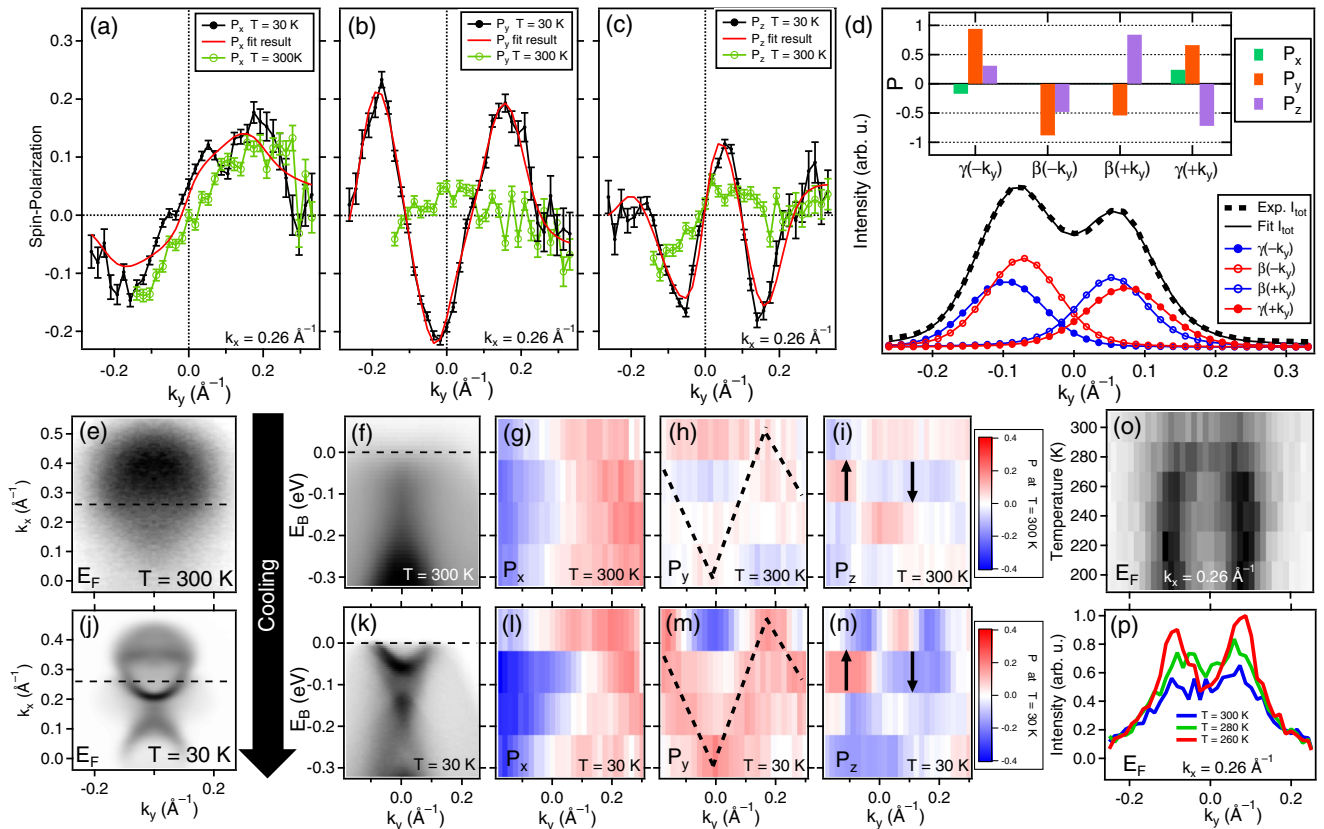


FIG. 3. (a)–(c) Spin-polarization momentum distribution curves at E_F and $k_x = 0.26 \text{ \AA}^{-1}$. (d) Results of vectorial spin analysis for the $T = 30 \text{ K}$ data, including peak intensities and spin components (inset). Temperature dependent measurements at (e)–(i) $T = 300 \text{ K}$ and (j)–(n) $T = 30 \text{ K}$. (e) and (j) Fermi surfaces. (f) and (k) ARPES intensity in gray scale and spin polarizations in a false-color (see inset) scale for (g) and (l) P_x , (h) and (m) P_y , and (i) and (n) P_z mapped over $E_B(k_y)$ at $k_x = 0.26 \text{ \AA}^{-1}$. All spin-resolved data were collected using 20 eV photons from the same sample, which was cleaved and measured at 300 K and then cooled. (o) and (p) Temperature dependence of high-resolution ARPES intensity at E_F and $k_x = 0.26 \text{ \AA}^{-1}$ collected using 67 eV photons for a sample cleaved at 300 K.

seen in Fig. 3(g). At both temperatures, hole-like states contribute an M shape of $+y$ -oriented spin in the energy-momentum maps of Figs. 3(h) and 3(m), as indicated by dashed lines, and z polarization that switches across $k_y = 0$ around $E_B = -0.1$ eV, as indicated by arrows in Figs. 3(i) and 3(n). This serves as a faint signature of T_d order persisting at 300 K.

Spin-integrated MDCs in Figs. 3(o) and 3(p) show the development of intensity in β upon cooling from 300 K, measured at E_F along the same momentum cut as in Figs. 3(a)–3(c). The peak intensities rise upon cooling from 300 to 280 K, but do not sharpen into clear, Lorentzian shapes until 260 K is reached. One could say that β either lies above E_F , is fully absent, or the signal is too broad and suppressed to be clearly observed at 300 K. ARPES spectra along $\bar{\Gamma}\bar{X}$ are shown divided by the Fermi cutoff in Figs. 4(a) and 4(b). For the case of a fresh surface prepared at 300 K shown in Fig. 4(a), β is not visible. It is shown elsewhere that, as in Figs. 3(o) and 3(p), β does not clearly emerge in this momentum cut either until the sample is cooled to 260 K [45]. Figure 4(b) shows that β , which presents a line of intensity connecting the bulk electron and hole states (green arrow) [27,45], persists after cooling to 120 K and annealing to 340 K. It is shown elsewhere that the chemical potential irreversibly increases by about 30 meV upon cooling through T_S [45]. It is likely that the spectral function of β is broadened and suppressed by scattering in the initial condition, obscuring the signal. These effects may have been diminished by the binding energy shift and/or improved structural order after one thermal cycle. The signal is simply not clear enough in the initial condition for further determination.

The signal of δ , indicated by red arrows in Figs. 4(a) and 4(b), is clear at certain emission angles (negative k_x) for

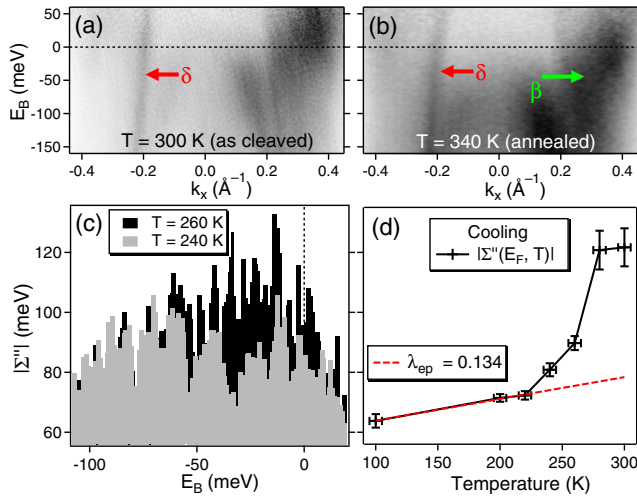


FIG. 4. (a)–(b) ARPES intensity along k_x at $k_y = 0$ divided by the Fermi cutoff. (c) Energy distribution curves of $|\Sigma''|$ in state δ at $k_y = 0$ for different temperatures as extracted from raw data. (d) Temperature dependence of $|\Sigma''(E_F)|$.

this case where p -polarized 67 eV photons are used. The steep hole-like dispersion reaches above $E_B = 50$ meV, which is around the maximum energy expected for WPs [28]. Additional measurements show that δ corresponds to what Ref. [23] referred to as a candidate topological surface state [45]. To investigate the response of electronic coherence to cooling, the magnitude of the imaginary part of the photohole self-energy $|\Sigma''|$ was computed by multiplying the group velocity with the peak half-width, using raw ARPES data collected at different temperatures [45]. There is a significant effect of noise on the results, but it can be appreciated from Fig. 4(c) that there is more area under the distribution of $|\Sigma''(E_B)|$ in the range $-50 \text{ meV} < E_B < 0 \text{ meV}$ at 260 K (black bars) than at 240 K (gray bars). Of the possible scattering mechanisms, only electron-phonon coupling (EPC) is expected to cause significant variation in $|\Sigma''(E_B)|$ near E_F [73]. In most metals, the lifetime broadening at E_F is proportional to the EPC constant λ_{ep} as $|\Sigma''(E_F, T)| = 2\pi k_B \lambda_{ep} T$, where k_B is the Boltzmann constant [74]. The average of broadening values extracted from the range $E_F \pm k_B T/2$ is shown as $|\Sigma''(E_F, T)|$ in Fig. 4(d), with the standard error of the mean shown as error bars. A linear fit in the 220–100 K region obtains a weak dependence on temperature corresponding to $\lambda_{ep} \approx 0.1$ plotted as a red dashed line in Fig. 4(d). Linear fitting in the 280–220 K region is unphysical ($|\Sigma''(E_F, T = 0)| < 0$). There is a rapid change in EPC, or at least some form of scattering, upon cooling through T_S . The EPC strength in $1T'$ and T_d -MoTe₂ is similar [5,9], but new forms of electron-lattice interaction arise in the case of strong disorder. For example, electron-phonon-impurity scattering [75,76], wherein electron-impurity and electron-phonon scattered paths interfere, can significantly contribute to the self-energy, even at high temperatures [77].

Spin polarization measured at $T > T_S$ could derive from a so-called hidden spin texture [78] that must exist in bulk $1T'$ -MoTe₂ because centrosymmetry is absent in all of the lattice sites [45]. However, a case of global $1T'$ order does not explain the anomalous lifetime broadening and one would expect a full lattice transition to produce a qualitative change in the measured spin-orientations that does not appear [45]. On the other hand, coexistence of $1T'$ and T_d structural phases would cause electrons to exist in mixed (noncoherent) states due to entanglement with variations in the lattice polarity, thus decreasing quasiparticle lifetimes and spin polarization. This is analogous to polar instability (PI) in ferroelectrics, wherein the symmetry-breaking order is short ranged or fluctuates. Sakai *et al.* [3] showed that PI exists in bulk $\text{Mo}_{1-x}\text{Nb}_x\text{Te}_2$ over a wide temperature range reaching up to 300 K beginning at $x = 0.08$, whereas it exists only in a narrow range around T_S for $x = 0$.

The surface sensitivity of (S)ARPES provides a means by which signatures of PI are observed around 300 K in the present experiments. Noting that increased electron density stabilizes the T_d structure [79,80], the surface dipole could

stabilize local T_d order at temperatures well above T_S , in analogy to so-called negative dead layers in ferroelectric materials [81]. Motion of the $T_d/1T'$ phase boundary is determined by the c -axis thermal gradient [4], which is well defined for a cooled sample with an exposed surface. It follows that the local electronic coherence improves as the boundary progresses into the bulk upon cooling. From the perspective of a surface-sensitive probe, the transition occurs gradually over a broader temperature range than in the bulk. Similarly, a gradual transition taking place around phase boundaries was found to coexist with a first-order bulk transition in NdNiO_3 , wherein boundary and bulk electronic properties were observed to evolve differently with temperature [82]. $\text{Mo}_{1-x}\text{Nb}_x\text{Te}_2$ alloys may lie in another extreme where, in the thermodynamic limit, a highly mixed bulk structure appears to change continuously with temperature, smoothing out the evolution of macroscopic transport properties [3].

In summary, the observed response to cooling the $1T'$ - MoTe_2 crystal is a gain in electronic coherence that yields a clear view of Fermi arcs and the novel 3D spin texture of T_d - MoTe_2 . The existence of finite P_z must be considered in future discussions of the magnetoresistance properties for T_d -(Mo/W) Te_2 materials. Both small and large Fermi arc states are observed at 340 K, where the volume of the bulk is almost entirely in the $1T'$ structural phase [2]. Therefore, the existence of the Fermi arcs is independent of any global, bulk Weyl semimetal phase of matter. Precise determination of the crystal structure near the surface (e.g., by scanning transmission electron microscopy) is vital for clarifying the relationship between the Fermi arcs and the Weyl and structural phases, the anomalous changes in self-energy broadening, and the origin of the spin texture observed at 300 K.

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