Switching of band inversion and topological surface states by charge density wave

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Topologically nontrivial materials host protected edge states associated with the bulk band inversion through the bulk-edge correspondence. Manipulating such edge states is highly desired for developing new functions and devices practically using their dissipation-less nature and spin-momentum locking. Here we introduce a transition-metal dichalcogenide VTe 2 , that hosts a charge density wave (CDW) coupled with the band inversion involving V3d and Te5p orbitals. Spin- and angle-resolved photoemission spectroscopy with first-principles calculations reveal the huge anisotropic modification of the bulk electronic structure by the CDW formation, accompanying the selective disappearance of Dirac-type spin-polarized topological surface states that exist in the normal state. Thorough three dimensional investigation of bulk states indicates that the corresponding band inversion at the Brillouin zone boundary dissolves upon the CDW formation, by transforming into anomalous flat bands. Our finding provides a new insight to the topological manipulation of matters by utilizing CDWs' flexible characters to external stimuli.
Since the discovery of topological insulators, a wide variety of topological phases have been intensively developed and established in realistic materials\(^1\). The upcoming target is to explore the guiding principles for the manipulation of these topological states. A key parameter to characterize the topological nature of materials is the band inversion realized by the crossing and anti-crossing of energy bands with opposite parities. In bulk materials, the control of band inversion has been mostly done through the tuning of spin-orbit coupling (SOC) by element substitution\(^3,4\), with some exceptions related to the topological crystalline insulators\(^5\). In the present work, we focus on charge density wave (CDW), i.e., the spontaneous modulation of charge density and lattice that modifies the periodicity and symmetry of the host crystal. In TaS\(_2\), for example, the Star-of-David type CDW superstructure induces the effective narrowing of valence bands thus causing the Mott transition\(^6,8\). From this viewpoint, CDW could also modify the band structures that involve the band inversion, and induce topological change. Moreover, it is worth noting that CDW can be flexibly controlled by external stimuli. In transition metal dichalcogenides (TMDCs), one of the well-known families of layered materials that host variety of CDWs reflecting their quasi-two dimensionality\(^4\). The transition metal dichalcogenides (TMDCs) are a well-known family of layered materials that host variety of CDWs reflecting their quasi-two dimensionality\(^9\). Manipulations of CDW states have been intensively investigated and realized, especially in the aforementioned archetypical material TaS\(_2\), by various stimuli, such as pressure\(^7\), electric field\(^10\), and optical pulse\(^11,12\). The feasibility of CDW thus paves the way toward triggering the exotic phase transitions and generating new functions. More recently, there has been increasing interest in SOC effect in TMDs\(^13\). Especially in the tellurides, (Mo,W)Te\(_2\)\(^14,16\) and (Ta,Nb)IrTe\(_2\)\(^17,18\) are reported to be topological Weyl semimetals. Here, the peculiar quasi-one-dimensional chain-like structures inherent to tellurides, as well as their stronger SOC compared with selenides and sulfides, are essential in realizing the topologically non-trivial states. In this stream, we focus on the telluride material VTe\(_2\), to investigate the interplay of band topology and CDW instability.

VTe\(_2\) has CdI\(_2\) structure in the high-temperature normal 1T phase, consisting of trigonal layers formed by edge-sharing VTe\(_6\) octahedra (Fig. 1a). With cooling, it undergoes a phase transition to the CDW phase at around 475 K, appearing as a jump in the temperature-dependent resistivity\(^19\). The resulting CDW state exhibits a (3 x 1 x 3) superstructure characterized by double zig-zag chains of vanadium atoms (Fig. 1d; hereafter we refer it as 1\(\times\)3\(\times\)3). This superstructure commonly appears in group-V transition metal ditellurides\(^3\), with some exceptions related to the topological states. A key parameter to characterize the topological CDW superstructure induces the effective narrowing of valence bands thus causing the Mott transition\(^6,8\). From this viewpoint, CDW could also modify the band structures that involve the band inversion, and induce topological change. Moreover, it is worth noting that CDW can be flexibly controlled by external stimuli. In transition metal dichalcogenides (TMDCs), one of the well-known families of layered materials that host variety of CDWs reflecting their quasi-two dimensionality\(^4\). The transition metal dichalcogenides (TMDCs) are a well-known family of layered materials that host variety of CDWs reflecting their quasi-two dimensionality\(^9\). Manipulations of CDW states have been intensively investigated and realized, especially in the aforementioned archetypical material TaS\(_2\), by various stimuli, such as pressure\(^7\), electric field\(^10\), and optical pulse\(^11,12\). The feasibility of CDW thus paves the way toward triggering the exotic phase transitions and generating new functions. More recently, there has been increasing interest in SOC effect in TMDs\(^13\). Especially in the tellurides, (Mo,W)Te\(_2\)\(^14,16\) and (Ta,Nb)IrTe\(_2\)\(^17,18\) are reported to be topological Weyl semimetals. Here, the peculiar quasi-one-dimensional chain-like structures inherent to tellurides, as well as their stronger SOC compared with selenides and sulfides, are essential in realizing the topologically non-trivial states. In this stream, we focus on the telluride material VTe\(_2\), to investigate the interplay of band topology and CDW instability.

Overviewing the electronic structures and modifications by CDW. Let us start by briefly overviewing the anisotropic modification of electronic structures from 1T to 1T\(\text{''}\) by presenting the ARPES data successfully focused on a CDW single-domain region. Figure 2a shows the ARPES image for the normal-state 1T-V\(_{0.87}S\text{Ta}_{0.13}Te\(_2\) taken at 300 K with a He-discharge lamp (photon energy \(h\nu = 21.2\) eV). We find V-shaped band dispersions along K – M – K that clearly cross the Fermi level \(E_F\). Zooming in the higher binding energy, we observe V-shaped bands that connect to the Dirac cone-like bands reminiscent of surface states in topological insulators, with the band crossing (Dirac points) at M. The topological character of these Dirac bands will be discussed later. On the other hand, Fig. 2b displays the ARPES results on a single-domain CDW state in 1T\(\text{''}\)-VTe\(_2\) (200 K, synchrotron light \(h\nu = 90\) eV). Because of the zigzag type CDW formation, the system now loses the threefold rotational symmetry, and the three equivalent M points in 1T turn into one M\(_1\) and two M\(_2\). Here, the V-shaped band and Dirac-like bands remain at M\(_1\), whereas at M\(_2\) side the unusual flat band is observed and the Dirac-like state is vanished. Thus, the 1T-1T\(\text{''}\) CDW transition induces the huge directional change of electronic structure accompanying the selective disappearance of Dirac-like states. In the following, we discuss these band structures in detail, by comparing with band calculations.

Results

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In this Article, we investigate the electronic structures of VTe\(_2\), by employing spin- and angle-resolved photoemission spectroscopy (ARPES) and first-principles calculations. We focus on the modifications of the bulk bands through the CDW formation, and its relevance to the Dirac surface states stemming from the topological band inversion. Experimentally it is not easy to perform highly precise ARPES measurement on the normal 1T phase of pristine VTe\(_2\) (>475 K), since such a high temperature may cause the sample degradation. Therefore we start from VTi\(_2\) showing a simple 1T structure down to the lowest temperatures\(^32\), and develop the single crystalline V\(_{1-x}\text{Ti}_{x}Te\(_2\) to access both 1T and 1T\(\text{''}\) phases at appropriate temperatures. Figure 1f displays the schematic electronic phase diagram of V\(_{1-x}\text{Ti}_{x}Te\(_2\), based on the temperature-dependent ARPES measurements. With increasing Ti, the CDW phase transition becomes gradually suppressed to the lower temperature region (see Supplementary Note 2). For investigating both the normal and CDW phases within a single sample, we synthesized the minimally Ti-doped single crystals (0.10 ≤ x ≤ 0.13) showing the transition just below room temperature (280–250 K). To discuss the electronic structures in momentum space, we introduce the following notation of the Brillouin zones (BZ). In 1T, the 2-dimensional (3-dimensional) BZ is represented by the hexagonal plane (prism) reflecting the trigonal symmetry (Fig. 1b, c). In 1T\(\text{''}\), the BZ changes into a smaller one of lower symmetry as indicated in Fig. 1e. In this paper, we use the 1T BZ notation to present the band structures in 1T and 1T\(\text{''}\) in a common fashion. To account for the in-plane anisotropy of vanadium chains that is essential in 1T\(\text{''}\), we define \(K_1\), \(K_2\) and \(M_1\), \(M_2\) as depicted in Fig. 1e (see Supplementary Note 1 for the details of BZ).

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To grasp the essential electronic modification via the CDW formation, we survey the temperature- and doping-dependent ARPES results (He-discharge lamp, $h\nu = 21.2$ eV). Note that the $1T^\prime$ phase inevitably contains the in-plane 120-degree CDW domains reflecting the threefold symmetry of 1T, and usually ARPES measurements include the signals of multiple domains (see Supplementary Note 5). Figure 3e, f respectively shows the ARPES image and corresponding EDCs of $V_{0.90}Ti_{0.10}Te_2$ in the CDW $1T^\prime$ phase. The blue broken lines highlight the shortest V-V bonds forming the double zigzag chains. The conventional unit cell is indicated by the black broken curve. The appearance of this anomalous flat band is thus the common signature of the CDW $1T^\prime$ phase, which however is seemingly beyond the simple band folding and gap opening in the Fermi surface nesting scenario. The localized nature of this electronic structure will be discussed later.

**Bulk and surface band structures in 1T normal phase.** Here we introduce the topological aspect that can be relevant to the 1T-1T$^\prime$ CDW transition. In the normal 1T phase, the band calculation suggests the band inversion involving V3$d$ and Te5$p$ orbitals at around the M and L points. The calculations of $1T-V_{0.87}Ti_{0.13}Te_2$ at several $k_z$, plotted along the direction parallel to $\Gamma$-M ($k_{3M}$, see Fig. 4a) are displayed in Fig. 4b. The colors of curves show the weight of atomic orbitals depicted by a false color-scale (see Supplementary Note 3 for detailed orbital components), whereas the black broken curves are the results without SOC. Focusing on the topmost two bands at M and L, labelled as A and B, we find that their orbital characters of mainly V3$d$ (blue-like, even parity) and Te5$p_x + p_y$ (red-like, odd parity) get inverted at a finite $k_z$. 

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**Fig. 1 Normal (1T) and CDW (1T$^\prime$) phases in VTe$_2$ system.** a Top view of VTe$_2$ layer in the high-temperature normal 1T phase. The conventional unit cell is indicated by the black lines. b (0 0 1) surface BZ of 1T-VTe$_2$. c Bulk BZ of 1T-VTe$_2$. d Top view of VTe$_2$ layer in the low-temperature CDW 1T$^\prime$ phase. The blue broken lines highlight the shortest V-V bonds forming the double zigzag chains. e (0 0 1) surface BZ considering the CDW in 1T$\prime$-VTe$_2$, superimposed on that in the 1T phase (dashed hexagon). f Schematic electronic phase diagram for $V_{1-x}Ti_xTe_2$, based on the temperature-dependent ARPES measurements.

**Fig. 2 Overview of Fermi surface and band dispersions for normal 1T and CDW 1T$^\prime$ phases.** a Bird’s eye ARPES image of 1T-$V_{0.87}Ti_{0.13}Te_2$ (300 K), $h\nu = 21.2$ eV). b Same as a, but for 1T$\prime$-VTe$_2$ (200 K, $h\nu = 90$ eV).
image of normal-state $V_{0.90}T_{0.10}Te_2$ along $\bar{K} - \bar{M} - \bar{K}$ (350 K, $h\nu = 21.2$ eV). By carefully analyzing the EDC/MDC (see Supplementary Note 7), we can indeed quantify the bottom of bulk band A ($E_A \sim 0.30$ eV), the top of bulk band B ($-0.90$ eV), and the crossing point of the surface Dirac cone (DP, $-0.66$ eV). We note that this bulk band A forms the triangular Fermi surfaces centered at $K$ points in the $k_z = 0$ plane, as shown in the Fermi surface image in Fig. 4f ($V_{0.87}T_{0.13}Te_2$, 300 K, $h\nu = 83$ eV).

We further perform the $h\nu$-dependent ARPES to confirm the two-dimensionality of the Dirac surface states and to clarify the $k_z$-dependent bulk electronic structure that is relevant to the band inversion. Figure 4h–j displays the ARPES images of $1\Gamma - V_{0.90}T_{0.10}Te_2$ near the M point (see the red arrow in Fig. 4g), recorded at 320 K with different photon energies, $h\nu = 63, 69$, and 78 eV, respectively corresponding to $k_z \sim 0$, $\pi/2c$, and $\pi/c$. Circle markers with vertical (horizontal) error bars represent the peak positions of EDCs (MDCs) (see Supplementary Note 7). Figure 4k displays the schematic band dispersions overlaid with the experimental peak plots extracted from Fig. 4h–j. Here we find that the bulk bands A and B clearly show the finite $k_z$-dispersions (respectively larger than 0.1 and 0.4 eV at M). As can be seen in Fig. 4b, this $k_z$-dispersion is essential for the band inversion at $(\pi/2c, 0, k_z)$, the origin of the topological surface state appearing around the M point. On the other hand, the Dirac surface state, that is highlighted by the overlaid orange curves in Fig. 4k, is almost independent of $h\nu$, indicating the two-dimensional nature of the topological surface state.

**Bulk and surface band structures in 17° CDW phase.** To unambiguously elucidate the anisotropic electronic structures in the CDW 17° phase, here we utilize the small spot size (typically 300 μm) of the synchrotron light beam and separately measure the in-plane CDW domains of VTe2 (see Supplementary Note 5). Figure 5a shows the Fermi surface image of the single-domain 17°-VTe2 (200 K, $h\nu = 90$ eV). We find that the two sides of the triangular Fermi surface around K observed in the normal 1T phase (Fig. 4f) are completely absent in the CDW state, and the remaining one forms the quasi-one-dimensional Fermi surface marked by the red broken curves. Figure 5b, d displays the ARPES images along representative two cuts (cut #1 and #2 as denoted in Fig. 5a) of the CDW phase (20 K, #2) for the same momentum region as c. The red and blue curves indicate the flat bands and dispersive band near the Fermi level, obtained in a similar manner with e, f. The red (blue) and purple (cyan) triangle markers respectively represent the peak positions of spin-up (down) spectra at $M_1$ and $M_2$ sides. As shown in the spin-resolved spectra in Fig. 5h, the lower branch of the Dirac cone clearly shows the spin polarization with sign reversal at (0 0 1) slab calculation (Fig. 4c), a new band dispersing around $E - E_p = -0.6$ eV near M can be recognized, that does not exist in the bulk calculations. It roughly follows the trajectory of the virtual crossing points of bulk bands A and B for no SOC. This is a surface state topologically protected by the band inversion at $(\pi/2, 0, k_z)$ occurring due to the moderate $k_z$ dispersions of V3d and Te5p bands. Indeed, in the $K - M - K$ direction (Fig. 4d), this surface state shows a Dirac cone-like dispersion connecting bands A and B. Figure 4e shows the ARPES

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**Fig. 3 Temperature/Ti-doping dependence of electronic structures.** a The band calculation of $V_{0.90}T_{0.10}Te_2$ in the normal phase along the high symmetry line (Γ–K–M–Γ). b The corresponding Fermi surface in the $k_z = 0$ plane. c ARPES image of $V_{0.90}T_{0.10}Te_2$ in the normal phase (350 K, marked as #1 in Fig. 1f) recorded on the high symmetry lines. The data are divided by the Fermi-Dirac function convoluted with the Gaussian resolution function, to eliminate the thermal broadening of the Fermi cutoff. The broken black curves are the guide for the band dispersions, obtained by tracking the peaks of the energy and momentum distribution curves. d Energy distribution curves along the M – K line for c (integral width: 0.05 Å$^{-1}$). e ARPES image of $V_{0.90}T_{0.10}Te_2$ in the CDW phase (20 K, #2) for the same momentum region as c. The red and blue curves indicate the flat bands and dispersive band near the Fermi level, obtained in a similar manner with c, f. Same as d, but for e and h. Same as e, f, but for the pristine VTe2 in the CDW phase (15 K, #3). All the ARPES data in the figure were obtained using a He-discharge lamp ($h\nu = 212$ eV). The data in e, h include the signal from all the in-plane 120-degree CDW domains.

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(0.76 π/c). In the corresponding (0 0 1) slab calculation (Fig. 4c), a new band dispersing around $E - E_p = -0.6$ eV near M can be recognized, that does not exist in the bulk calculations. It roughly follows the trajectory of the virtual crossing points of bulk bands A and B for no SOC. This is a surface state topologically protected by the band inversion at $(\pi/2, 0, k_z)$ occurring due to the moderate $k_z$ dispersions of V3d and Te5p bands. Indeed, in the K – M – K direction (Fig. 4d), this surface state shows a Dirac cone-like dispersion connecting bands A and B. Figure 4e shows the ARPES
Fig. 4 Topological character of band structures in the normal 1T phase for Ti-doped VTe2. a The Brillouin zone of 1T phase. kx, ky, and kz indicate the axes in the reciprocal space corresponding with the global xyz axes (see Fig. 1a) used for the calculation in b. b The bulk band calculations of 1T-V_{0.87}Ti_{0.13}Te$_2$ along k$_{\Gamma M}$ direction at several k$_z$ values (0, 0.38, 0.76, 1.00 Å/$c$). The color of the curves indicates the weight of orbital characters, as shown by the color-scale. The “+” and “−” mark the even and odd parity, respectively. The broken curves represent the results without spin-orbit coupling. c, d The corresponding (0 0 1) slab calculations along the Γ–M (c) and the K–M–K (d) lines, respectively. e ARPES image of 1T-V$_{0.91}$Te$_{0.09}$Te$_2$ along the K–M–K direction recorded using a He-discharge lamp (21.2 eV, 350 K). The red (blue) marks represent the peak positions of energy (momentum) distribution curves. f Fermi surface image of 1T-V$_{0.87}$Ti$_{0.13}$Te$_2$ in the k$_z$ = 0 plane (circular polarized 83 eV photons, 300 K), with an energy window of ±5 meV. The red curves display the Fermi surface schematically. g Schematic 2D BZ. The red arrow indicates the measurement region of hv-dependent ARPES. h–j hv-dependent ARPES spectra of 1T-V$_{0.90}$Te$_{0.10}$Te$_2$ with 63 eV (h), 69 eV (i), and 78 eV (j) photons (circular polarization, 320 K), which respectively detects at k$_z$ = 0, π/2c, and π/c plane. The circles with vertical (horizontal) error bars represent the EDC’s (MDC’s) peak positions. k Schematic band dispersion along K – M – K with experimentally obtained peak plots. The red, green, and blue markers are the peak plots for hv = 63, 69, and 78 eV, respectively from h–j. The orange curve represents the schematic of the Dirac surface state (DSS), whereas the blurred gray curves are the bulk bands (Bulk A and B).

M$_1$ point (i.e., Dirac crossing point), similarly to the topological surface state in the topological insulators. On the other hand, the bulk flat bands around $E_B = 0.25$ eV at M$_1$ side indeed show the spin degenerate character. Figure 5i is an expanded viewgraph of spin-resolved spectra near the Fermi level at emission angle $\theta = \pm 4^\circ$. There are slight spin-up/down intensity contrasts just below $E_F$ ($E_B = 0.05$ eV), which should be corresponding to the upper branch of the surface Dirac cone band at M$_1$ side.

Similarly to the normal phase, hv-dependent ARPES measurement is performed on the CDW state VTe$_2$ (15 K, multi-domain) to clarify the $k_z$-dependence of electronic structures. Figure 5j–l displays the ARPES image near the M$_1$/M$_2$ points (see the red arrow in Fig. 5f), recorded with hv = 54, 61.5, and 69 eV, respectively. Again by comparing with the single-domain measurement, we can assign the peaks to either M$_1$ or M$_2$ side (see Supplementary Note 9).
the schematic band dispersions along $K_1 - M_1 - K_1$ and $K_1 - M_2 - K_2$, overlaid with the experimental peak plots extracted from Fig. 5j–l. At the $M_1$ side, the V-shaped band with high intensity clearly shows the finite $k_z$-dispersion of $>0.2$ eV, together with the $h$-independent Dirac surface state. These are similar to the case of normal 1T phase along $K - M - K$ (Fig. 4k). On the other hand, at the $M_2$ side, the emergent flat band shows negligible $k_z$-dependence, in contrast to the V-shaped band observed at the $M_1$ side. With this we can conclude that the electronic state at the $M_2$ side has unusually localized nature, which should give rise to the dissolution of the band inversion along $k_z$ direction.

**Picture of CDW state based on orbital bonding.** To grasp the essential feature of the CDW state, here we introduce the local picture. For simplicity, we adopt an orthogonal octahedral
XYZ coordination by considering the VTe$_6$ octahedron as shown in Fig. 6a, and focus on V3d t$_{2g}$ (d$_{xy}$, d$_{yz}$, d$_{zx}$) orbitals that dominate the density of states near $E_F$ (see Supplementary Note 10). Note that this is different from the global $xyz$ setting adopted in Fig. 4b, where $z$ corresponds to the stacking direction. Here we choose $Z$ as the V-Te bond direction that is perpendicular to the vanadium’s chain direction $b_n$.

Figure 6b shows the calculated partial density of states (PDOS) for vanadium d$_{xy}$, d$_{yz}$, and d$_{zx}$ orbitals. In 1$T$ (Fig. 6b left), they are naturally degenerate reflecting the trigonal symmetry. In 1$T''$ (Fig. 6b right), on the other hand, d$_{yz}$/d$_{zx}$ and d$_{xy}$ orbitals have strikingly different distributions. We can classify the PDOS for 1$T''$ into three major parts, lower, middle, and upper, respectively lying around $E_F$ $\sim$ 0.4, $-$0.5, and $-$1.3 eV. They are indicative of bonding (B), nonbonding (NB), and antibonding (AB) bands, arising from the trimerization-like displacements of three adjacent vanadium atoms formed by the $\sigma$-bonding of d$_{yz}$/d$_{zx}$ orbitals\textsuperscript{23,24}, as marked by the pink oval in Fig. 6a for d$_{yz}$. Through the d$_{yz}$/d$_{zx}$ trimerization, the remaining d$_{xy}$ stays relatively intact, thus its PDOS mostly contributes to the middle band. Here, the d$_{yz}$/d$_{zx}$ bonding band makes a peak in PDOS at around $E_F$ $\sim$ 0.3 eV, corresponding to the flat bands lying around $\Gamma$ $-$ K$_2$ $-$ M$_2$ (see Supplementary Note 10 for the band dispersions). The experimentally observed flat band in Fig. 5d should be thus reflecting the localized nature of d$_{yz}$/d$_{zx}$ trimers.

Such orbital bonding picture is also useful for the intuitive understanding of Fermi surfaces at BZ boundary. Figure 6c depicts the schematics of the Fermi surfaces with the locations of the Dirac crossing points (yellow circle markers at M points). According to the calculation in 1$T$, the three sides of the triangular Fermi surface mainly consist of d$_{yz}$, d$_{zx}$, and d$_{xy}$ orbitals, respectively (see Supplementary Note 10). This can be regarded as the virtual combination of one-dimensional Fermi surfaces formed by the $\sigma$-bonding of d$_{yz}$, d$_{zx}$, and d$_{xy}$ orbitals, raised as the basic concept of the hidden nesting scenario in CdI$_2$-type TMDCs\textsuperscript{23} and purple bronzes AMo$_6$O$_{12}$ (A = Na, K)\textsuperscript{25-27}. In the CDW 1$T''$ phase, the two sides of the Fermi surfaces composed of d$_{yz}$/d$_{zx}$ orbitals (M$_2$ sides) turn into the localized flat bands as a consequence of the vanadium trimerization, while the one composed of d$_{xy}$ remains intact. This explains the quasi-2D to quasi-1D change of the triangular Fermi surface at BZ boundary as obtained in the present result.

Discussion

Figure 6d summarizes the observed anisotropic reconstruction of the bulk and topological surface states occurring around the BZ boundaries by the CDW formation. As discussed, the band structures observed in the normal 1$T$ phase are basically in a good agreement with our calculations, indicating the triangular hole Fermi surfaces around the K points. At M points, the Dirac-type topological surface states exist in addition to the V-shaped bulk bands. We attribute this to the band inversion of V3d and Te5p orbitals occurring at ($a^*$/2, 0, k$_z$), which can be traced...
The electronic structures of (V,Ti)Te$_2$ by utilizing ARPES and selectively switching off the band inversion and corresponding further explained based on the change in crystal symmetry. In the other hand, while the curvature and topological invariants of materials should be determined by considering the detailed modiﬁcation, leading to the topological changes in particular bands. Considering that CDWs are often changeable to external stimuli, we can also expect to manipulate the CDW-coupled topological state. The effect of thinning the crystals down to atomic thickness is also worth investigating, which may give rise to hidden CDW states and electronic phase transitions. The combination of CDW ordering and topological aspects may lead to the new stage of manipulating the quantum materials.

Methods

Sample preparation. High-quality single crystals of V$_x$Ti$_{1-x}$Te$_2$ were grown by the chemical vapor transport method with TeCl$_2$ as a transport agent. Temperatures for source and growth zones are respectively set up as 600°C and 550°C. The Te concentrations (x) were characterized by energy dispersive x-ray spectrometry (EDX) measurements. To access both 17$^\text{th}$ and 17$^\text{th}$ phases, we used V$_{0.90}$Ti$_{0.10}$Te$_2$, V$_{0.97}$Te$_2$, and V$_{0.87}$Te$_2$ samples with the transition temperature of –280 K, –250 K and –475 K, respectively.

ARPES measurements. ARPES measurements were made at the Department of Applied Physics, The University of Tokyo, using a VUV5000 He-discharge lamp and a VG-Scienta R4000 electron analyzer. The photon energy and the energy resolution are set to 21.2 eV (Hel) and 15 meV. The spot size of measurements was ~2 × 2 mm$^2$. The data in Figs. 2a, 3c–h and 4e were obtained with this condition.

Photon-energy-dependent and domain-selective ARPES measurements were conducted at BL28A in Photon Factory (KEK) by using a system equipped with a Scienta SES2002 electron analyzer. Photon-energy-dependent measurements on 17$^\text{th}$- and 17$^\text{th}$-VTe$_2$ were performed respectively using circular and s-polarized light (hv = 45 – 90 eV). The energy resolution was set to 30 meV. The relation between the incident light energy and the detected $k_z$ value is estimated from the experimentally obtained work function (~3.7 eV) and inner potential (~8.8 eV). The data in Figs. 4b–j and 5j–l were obtained in this condition. Domain-selective measurements on 17$^\text{th}$- and 17$^\text{th}$-VTe$_2$ were performed at 200 K using a 90 eV circularly polarized light with a spot size of ~100 × 300 μm$^2$. The energy resolution was set to 50 meV. The data in Fig. 5b, d were obtained from the different in-plane domains of a single sample, by translating the sample without changing its orientation. The Fermi surface images in Fig. 4f, 5a were also obtained with this instrument.

Spin-resolved ARPES measurements were performed at Efficient Spin Resolved Spectroscopy end station attached to the APPLE-II-type variable polarization undulator beamline (BL-9B) at the Hiroshima Synchrotron Radiation Center (HSRC). The analyzer of this system consists of two sets of very-low-energy electron diffraction spin detectors, combined with a hemispherical electron analyzer (VG-Scienta RA4000). Measurements of 17$^\text{th}$-VTe$_2$ were performed using a s-polarized 21 eV light at 15 K. In the spin-resolved measurements, we set the energy and angle resolutions to 120 meV and ±1.5°, respectively. We adopted the Sherman function $S_{\text{Sherman}} = 0.25$ for analyzing the obtained data. The data in Fig. 3g–i were obtained with this condition.

In all the ARPES experiments, samples were cleaved at room temperature in UHV, and the vacuum level was better than 5 × 10$^{-10}$ Torr through the measurement. The Fermi level of the samples was referenced to that of polycrystalline golds electrically connected to the samples.

Band calculations. The relativistic electronic structure of 17$^\text{th}$-V$_{0.87}$Ti$_{0.13}$Te$_2$ was calculated within the density functional theory (DFT) using the Perdew-Burke-Ernzerhof (PBE) exchange-correlation functional corrected by the semilocal Tran-Behcke-Johnson potential, as implemented in the WFN3S package. The effect of Ti doping was treated within the virtual crystal approximation. The BS was sampled with a 20 × 20 × 20 k-mesh and the muffin-tin radius $R_{\text{MT}}$ for all atoms was chosen such that its product with the maximum modulus of reciprocal vectors $\mathbf{R}_{\text{MT}}$ becomes $R_{\text{MT}}\mathbf{R}_{\text{MT}} = 7.0$. To describe the surface electronic structure, the bulk DFT calculations were downfolded using maximally localized Wannier functions and the resulting 22-band tight-binding transfer integrals implemented within a 200-unit supercell. For the pristine 17$^\text{th}$- and 17$^\text{th}$-VTe$_2$, the DFT electronic structure calculations were carried out by OpenMX code (http://www.openmx-square.org/), using the PBE exchange-correlation functional and a fully relativistic j-dependent pseudopotentials. We adopted and fixed the crystalline structures of 17$^\text{th}$- and 17$^\text{th}$-VTe$_2$ reported in ref. 20, 41 and sampled the corresponding $B_z$ by a 8 × 8 × 8 k-mesh.

Data availability

The datasets that support the findings of the current study are available from the corresponding author on reasonable request.

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Author contributions

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Competing interests

The authors declare no competing interests.

Additional information

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