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Monolayer VTe₂: incommensurate Fermi-surface nesting and suppression of charge density wave

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Abstract

We investigated the electronic structure of monolayer VTe₂ grown on bilayer graphene by angle-resolved photoemission spectroscopy (ARPES). We found that monolayer VTe₂ takes the octahedral 1T structure in contrast to the monoclinic one in the bulk, as evidenced by the good agreement in the Fermi-surface topology between ARPES results and first-principles band calculations for octahedral monolayer 1T-VTe₂. We have revealed that monolayer 1T-VTe₂ at low temperature is characterized by a metallic state whereas the nesting condition is better than that of isostructural monolayer VSe₂ which undergoes a CDW transition to insulator at low temperature. The present result suggests an importance of Fermi-surface topology for characterizing the CDW properties of monolayer TMDs.

Layered transition-metal dichalcogenides (TMDs) are a promising candidate for realizing outstanding properties associated with two-dimensionalization since bulk TMDs are known to exhibit various physical properties such as magnetism, Mott-insulator phase, and charge density wave (CDW), besides the wide range of transport property (insulator, semiconductor, metal, and superconductor), most of which are prone to the change in the dimensionality of materials. When TMDs are thinned to a single monolayer (2D limit), they exhibit even more outstanding properties distinct from bulk, as represented by the room-temperature ferromagnetism in VSe_2 in contrast to the nonmagnetic nature of bulk¹ and the change in the band-gap property from indirect to direct transition in MoS_2 ². The CDW is a most pronounced phenomenon widely seen in both bulk and atomic-layer TMDs. In bulk TMDs, the interplay between the Fermi-surface nesting and the energy-gap opening as well as its relationship to the strength of CDW properties such as the CDW transition temperature (T_{CDW}) has been a target of intensive studies³.

The role of dimensionality to the mechanism of CDW, in particular whether or not the CDW is more stable in the 2D limit, is now becoming a target of fierce debates, being stimulated by a recent success in fabricating various atomic-layer TMDs by exfoliation and epitaxial techniques. Recent studies on some TMDs such as TiSe_2 , VSe_2 , and NbSe_2 ^{4–8} have shown a marked increase in T_{CDW} upon reducing the thickness down to a few monolayers. In contrast, it has been reported that the CDW vanishes in monolayer TaS_2 and TaSe_2 ^{9,10}, suggesting an important role of substrate and many-body effects. In $1T\text{-VSe}_2$, reducing the number of layers by exfoliating bulk crystal leads at first to gradual decrease of T_{CDW} , but at a critical film thickness of ~ 10 nm, the T_{CDW} exhibits a characteristic upturn and reaches 140 K in a few monolayers, much higher than bulk T_{CDW} (110 K)¹¹. Such enhancement of T_{CDW} in monolayer VSe_2 was also revealed by angle-resolved photoemission spectroscopy (ARPES) wherein the role of FS nesting and electron-phonon coupling was intensively debated^{4,6,7,12–14}. However, the nature of 2D CDW is still far from reaching a consensus, as highlighted by a wide variety of periodic lattice distortions hitherto proposed for monolayer VSe_2 (e.g., 4×4 , 4×1 , $\sqrt{7} \times \sqrt{3}$, $4 \times \sqrt{3}$)^{6,7,12–14}. In a broader perspective, it is still unclear to what extent the conventional FS-nesting picture can be applied to the 2D CDW materials and how the FS topology and electronic interactions are related to the CDW properties such as the enhancement/suppression of T_{CDW} . To address these essential questions, a further study on the electronic structure with new monolayer TMDs is highly

required.

In this Rapid Communication, we report the first successful fabrication of monolayer VTe_2 on bilayer graphene / SiC and its ARPES investigation. While the monoclinic ($1T'$) phase is known to be stable in bulk VTe_2 , our monolayer VTe_2 film takes the octahedral $1T$ structure [see Fig. 1(a)]. Importantly, this enables us to directly compare the electronic states and CDW properties with isostructural monolayer $1T$ - VSe_2 . Our ARPES study on monolayer VTe_2 signifies a large, nearly perfectly nested triangular FS centered at the K point in the Brillouin zone (BZ). We also found that the V $3d$ band apparently crosses the Fermi level (E_F) midway between the Γ and K points, indicative of the metallic state at low temperature, unlike monolayer VSe_2 which shows a fully gaped insulating state below 140 K. We discuss possible origins for such an intriguing difference in terms of the variation in the FS-nesting condition and electron-phonon coupling.

A high-quality monolayer VTe_2 film was grown on bilayer graphene by the molecular-beam-epitaxy (MBE) method. ARPES measurements were performed at the BL-28B beamline in Photon Factory, KEK. First-principles band-structure calculations were carried out using the QUANTUM ESPRESSO code with generalized gradient approximation^{15,16}. For details, see section 1 of Supplemental Material¹⁷.

First, we present characterization of monolayer VTe_2 film. Figure 1(b) shows the RHEED pattern of pristine bilayer graphene on SiC(0001) substrate. We clearly observe the 1×1 and $6\sqrt{3} \times 6\sqrt{3}$ $R30^\circ$ streak patterns, which correspond to bilayer graphene and underlying carbon-mesh layer on SiC, respectively¹⁸. After growing a VTe_2 film by co-depositing V and Te atoms onto the bilayer-graphene surface, the RHEED intensity from the substrate disappears, and a sharp 1×1 streak pattern appears [Fig. 1(c)], similarly to the case of other monolayer TMD films grown on bilayer graphene^{6,19,20}, indicating the formation of VTe_2 . Absence of additional streak patterns suggests no inclusion of monoclinic phase with zigzag chain structure which is known to exist in bulk VTe_2 below 482 K^{21,22}. This situation is particularly suited for comparing the electronic states with other $1T$ monolayer polymorphs. We come back to this point later in detail. As seen in the *ex-situ* AFM image in Fig. 1(d), large islands with a typical height of 0.8 nm, which corresponds to that of monolayer [Fig. 1(e)], are recognized on bilayer graphene. The energy distribution curve (EDC) at the Γ point in Fig. 1(f) signifies no detectable photon-energy ($h\nu$) variation in the energy position of bands, supporting the 2D nature of electronic states.

We have estimated the in-plane lattice constant of monolayer VTe₂ as $a \sim 3.35$ Å at room temperature by comparing the relative position of the RHEED patterns between graphene and VTe₂. This value is in good agreement with that estimated from the absolute wave vector values at the M (1.08 ± 0.03 Å⁻¹) and K (1.24 ± 0.03 Å⁻¹) points relative to the Γ point in the ARPES data (3.35 ± 0.09 Å). (including error bars due to the angular resolution and the angle-to- $\mathbf{k}$ conversion). Intriguingly, these values are much smaller than that of bulk octahedral 1T-VTe₂ (3.64 Å; obtained above 482 K where the 1T phase is stable). Taking into account that the coupling between graphene and 1T-VTe₂ film could be sufficiently weak due to the van-der-Waals-coupling nature, it is inferred that the value of 3.35 Å could be the most stable lattice parameter for free-standing monolayer VTe₂.

Figures 2(a) and 2(b) display the plot of valence-band ARPES intensity for monolayer VTe₂ measured at $T = 40$ K along the Γ M and Γ K cuts with $h\nu = 56$ eV, compared with the corresponding band dispersion obtained by the first-principles band-structure calculations for free-standing monolayer 1T-VTe₂ with the input of experimental lattice constant a . One can see in Fig. 2(a) several energy bands whose dispersion appears to be symmetric with respect to the Γ point. A side-by-side comparison of Figs. 2(a) and 2(b) also reveals a good agreement in the overall valence-band structure between the experiment and calculations, demonstrating that the fabricated monolayer VTe₂ indeed takes the 1T structure (see section 2 of Supplemental Material for details¹⁷). According to the calculations, energy bands lying at the binding energy (E_B) of 1-6 eV, including the holelike bands which rapidly move toward E_F on approaching the Γ point, are attributed to the Te 5p orbitals, while the energy band within 0.5 eV of E_F with a relatively flat dispersion around the M point is assigned as the V 3d band.

To see more clearly the electronic states responsible for the physical properties, we show in Figs. 2(c) and 2(d) the ARPES intensity near E_F at $T = 40$ K measured along the Γ M and Γ K cuts, respectively. A detailed spectral analysis by tracing the peak position of EDCs suggests that two topmost Te 5p bands with different band velocities do not reach E_F , but are topped at 60 meV below E_F at the Γ point. These bands are degenerate exactly at the Γ point, consistent with the calculation in Fig. 2(b). We found that the shallow V 3d band is also located at ~ 60 meV below E_F at the Γ point and disperses toward higher E_B on approaching the M point [Fig. 2(c)], while it crosses E_F midway between the Γ and K points, accompanied with a sudden drop in the spectral weight [Fig. 2(d)]. While the overall

experimental band structure shows a good agreement with the calculated band structure for the $1T$ phase, we found that some bands near E_F in the experiment are renormalized with respect to those in the calculations (for details, see section 2 of Supplemental Material¹⁷). It is noted here that we found no evidence for the energy splitting of bands associated with possible exchange splitting due to ferromagnetism, which is further corroborated by our x-ray magnetic circular dichroism measurement at 80 K showing no change in the V $L_{2,3}$ -absorption edge across the magnetic-field reversal²³. This suggests the absence of ferromagnetic order in monolayer VTe₂. At this stage, it is unclear why the ferromagnetism appears in monolayer VSe₂ but not in monolayer VTe₂, though it is noted that the ferromagnetism in monolayer VSe₂ itself is contradictory and is currently a target of fierce debate^{1,6,7}. It is also unknown whether or not the ferromagnetic property is related to the CDW.

To clarify the topology of FS, we have performed ARPES measurements in 2D $\mathbf{k}$ space. Figures 3(a)-3(c) show the contour maps of ARPES intensity for different E_B slices. At $E_B = E_F$ [Fig. 3(a)], one can recognize a couple of fairly straight intensity patterns around the M point running parallel to the ΓM direction (red dashed line). This intensity pattern forms a large, almost perfectly triangular-shaped FS enclosing the K point. Remarkably, this FS is well reproduced by the calculations for free-standing $1T$ -VTe₂ [Fig. 3(d)] with the input of experimental lattice constant, confirming again the $1T$ nature of our epitaxial film. Absence of any spurious intensity that could be associated with the band folding with (3×1) periodicity expected from the formation of double zigzag-chain superstructure seen in bulk VTe₂²² further corroborates the purely $1T$ nature of the film (see section 4 of Supplemental Material for details¹⁷).

Upon increasing E_B to 0.2 eV [Fig. 3(b)], the experimental triangular pattern seen at $E_B = 0$ eV [Fig. 3(a)] transforms into a M-point-centered ellipsoid elongated along the ΓM direction, which shrinks on further increasing E_B to 0.4 eV [Fig. 3(c)]. This indicates that the triangular FS forms a hole pocket, consistent with the calculated band dispersion in Fig. 2(b) in which the V $3d$ band is located at ~ 1 eV above E_F at the K point. Figure 3(e) shows the ARPES-derived band structure along three representative k cuts (cuts A-C) which cross the triangular FS. On cut A which touches the corner of triangular FS, one can see a couple of V-shaped bands in the vicinity of E_F . These two V-shaped bands are gradually separated from each other on going from cut A to cuts B and C, indicating that the triangular FS is holelike. In Figs. 3(b) and 3(c), one can also identify an intense circular

spot at the Γ point stemming from the Te 5*p* bands. We emphasize again that although the proximity of Te 5*p* bands to E_F enhances the intensity at the Γ point, these fully occupied bands do not participate in the FS. Therefore, the FS of monolayer VTe₂ is solely dictated by the triangular hole pocket at the K point, which greatly simplifies the discussion on the FS topology and nesting, as detailed later. We have estimated the total carrier concentration to be 0.98 ± 0.08 electrons / unit cell, by evaluating the area of FS with respect to that of whole BZ. This suggests that our monolayer film keeps stoichiometry and no observable charge transfer from the substrate takes place.

Now that the FS topology is established, we shall address a key question regarding a possible energy gap opening associated with the occurrence of CDW. We selected a $\mathbf{k}$ cut passing the corner of triangular FS [blue line in Fig. 4(a)], and show the ARPES intensity at $T = 40$ K plotted as a function of wave vector and E_B symmetrized with respect to E_F in Fig. 4(c). One can clearly see a dispersive band reaching E_F showing the brightest intensity at E_F , indicating the absence of an energy gap. This is in sharp contrast to the result of monolayer VSe₂ [Figs. 4(b) and 4(d)] that signifies a marked suppression of intensity within ± 0.1 eV of E_F at $T = 40$ K due to the CDW-gap opening⁶. To see the low-energy spectral feature in more detail, we have performed high-resolution ARPES measurements along several cuts crossing the FS, and show the EDCs at various k_F (Fermi wave vector) points (points 1-7) covering the whole straight segment of the triangular FS in the first BZ in Fig. 4(e). The corresponding symmetrized EDCs in Fig. 4(f) show a single peak at points 1 and 2 located around the corner of FS, while the EDCs at points 3-7 exhibit a weak dip structure at E_F indicative of a large residual spectral weight at E_F . We attribute this spectral-weight suppression as the pseudogap, but not the CDW gap, since the spectral behavior resembles that of monolayer VSe₂ at room temperature (well above $T_{CDW} \sim 140$ K), which shows the coexistence of pseudogap and metallic Fermi-arc states⁶. Also, the pseudogap of VTe₂ persists over a wide temperature range (10 - 300 K), similarly to VSe₂. It is noted that the pseudogap is unlikely to be due to some extrinsic effects such as the sample/surface quality and/or the experimental conditions (e.g. photoionization cross-section and light polarization), but is an intrinsic feature of monolayer VTe₂. The pseudogap may be explained in terms of the CDW fluctuations and/or the electron-phonon coupling associated with the CDW (see also section 3 of Supplemental Material¹⁷). The metallic state revealed in Figs. 4(c) and 4(e) in VTe₂ is obviously different from the fully gapped

insulating state below T_{CDW} , as visible from the strong spectral-weight suppression around E_F over the entire FS as seen in Fig. 4(g). We thus conclude that the CDW is suppressed in monolayer VTe_2 .

A key to understand such a contrasting behavior may lie on the difference in the FS topology between the two V-dichalcogenide monolayers. Figure 4(h) directly compares the FS obtained from ARPES measurements of monolayer VTe_2 and VSe_2 ⁶. One can immediately recognize that both monolayers show a similar triangular pocket at the K point whereas a circular hole pocket exists only in VSe_2 . Extra hole carriers at the Γ -centered pocket in VSe_2 resides on the K-centered pocket in VTe_2 , as seen from a larger triangular pocket in VTe_2 . This is reasonable since the Se and Te atoms are isovalent and the total FS area should be the same. The expansion of triangular pocket would widen the straight segment of the FS in VTe_2 . Assuming that the nesting vector q is parallel to the ΓM direction⁶, this would lead to an enhancement of electronic susceptibility in VTe_2 . Thus, one would naively expect that the CDW in VTe_2 is more stable than that in VSe_2 according merely to the FS-nesting picture. However, this is not the case since the CDW appears to be suppressed in VTe_2 . Thus, one cannot sufficiently describe the CDW of monolayer VSe_2 or VTe_2 simply in terms of the energy gain around E_F in the electronic system, which suggests the importance of considering the electron-phonon coupling²⁴. While detailed discussion on the relevance of electron-phonon coupling requires sophisticated first-principles band-structure calculations, we point out here a possibility that such an electron-phonon coupling could be linked to the electronic states via the commensurability of the nesting. We found that the nesting vector along the ΓM direction in VSe_2 is commensurate to the lattice ($1/4 \mathbf{G}$ where $\mathbf{G}$ is the reciprocal lattice vector)⁶, while that in VTe_2 is incommensurate ($1/4.6 \mathbf{G}$). If such commensurability enhances the electron-phonon coupling at the corresponding lattice periodicity, it may stabilize the CDW. However, this explanation is still speculative, requiring further experimental and theoretical studies to firmly pin down the CDW origin.

Our band calculations of monolayer VTe_2 show that a small (1.5 %) change in the in-plane lattice constant is sufficient to control the emergence/absence of a small hole pocket at Γ and the concomitant change in the FS-nesting condition at the triangular pocket. Such sensitivity of the FS topology to the lattice parameters is essential due to the fact that the narrow V $3d$ band is located in the vicinity of E_F . Therefore, we expect that small perturbations such as lattice strain and carrier doping would easily trigger the change in the

FS topology and the nesting vector, leading to the modulation of CDW properties. In this regard, the reported differences in the FS topology around the Γ point in VSe_2 , which may link to diverse periodic lattice distortions^{4,6,7,13}, may be interpreted in terms of a reflection of the high sensitivity of the CDW characteristics to the strain and carrier balance. The present result suggests an importance of precisely controlling the lattice strain and carrier concentration for manipulating the CDW. Such band engineering would be a main target of future studies.

In conclusion, we have performed an ARPES study on monolayer $1T\text{-VTe}_2$ grown on bilayer graphene by MBE. We found a large triangular FS at the K point that satisfies a nearly perfect nesting condition, whereas the CDW is suppressed as highlighted by the observation of E_F -crossing of bands at low temperature, in contrast to monolayer VSe_2 that exhibits a well-defined CDW characterized by the fully gapped insulating state. The present result opens a pathway toward controlling novel physical properties of 2D TMDs through the band engineering.

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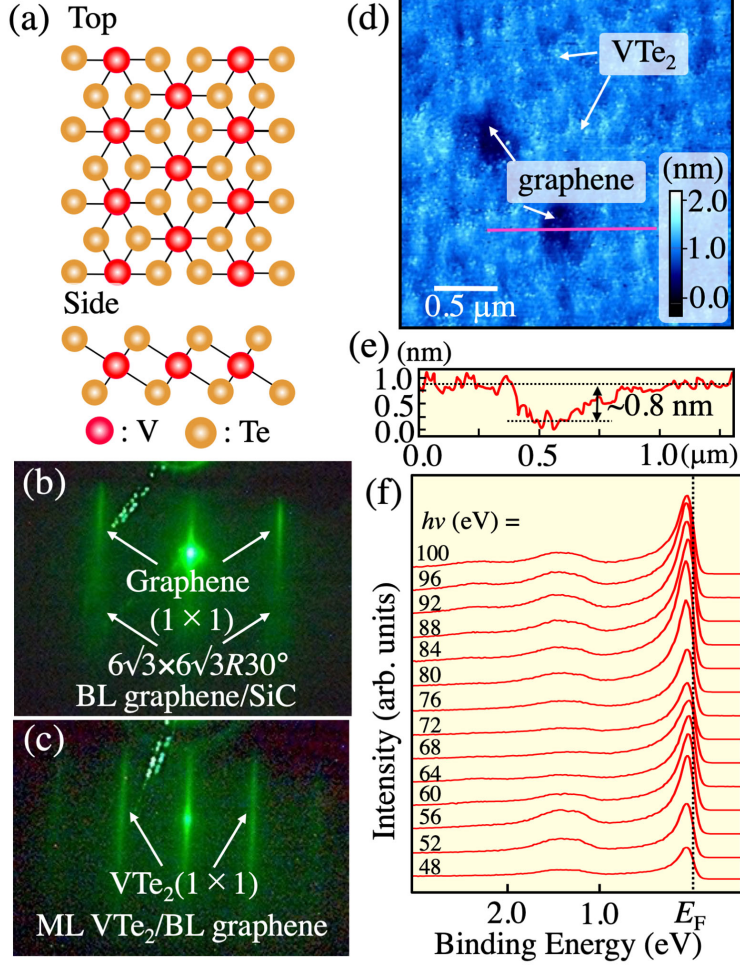


FIG. 1. (a) Crystal structure of monolayer 1T-VTe₂. (b),(c) RHEED patterns of bilayer (BL) graphene and monolayer (ML) VTe₂ on BL graphene, respectively. (d) AFM image of monolayer VTe₂. High (white), medium (blue), and low (dark) intensity regions are attributed to atoms/molecules adsorbed on VTe₂, clean monolayer VTe₂, and atoms molecules adsorbed on graphene substrate, respectively. The reason why we did not attribute the dominant white and blue areas to graphene or 2 monolayer VTe₂ is because we performed *in-situ* ARPES measurements with the same sample and observed a dominant contribution from the monolayer energy bands to the total ARPES intensity. (e) Height profile along a cut shown by magenta solid line in (d). (f) Photon-energy dependence of the EDC at the Γ point in monolayer VTe₂.

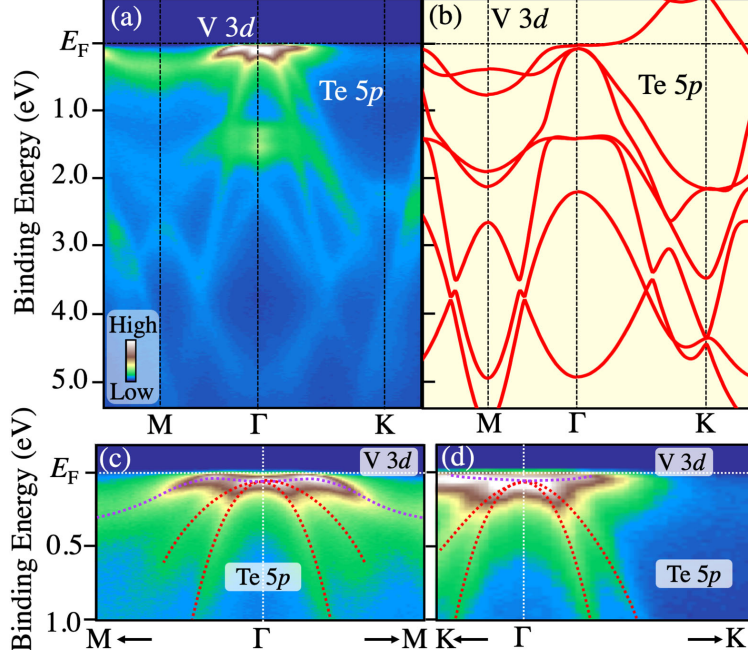


FIG. 2. (a) Plot of ARPES intensity for monolayer VTe_2 along the Γ M and Γ K cuts measured with $h\nu = 56$ eV at $T = 40$ K. (b) Band structure obtained from the first-principles band-structure calculations for monolayer $1T$ - VTe_2 with the input of experimental in-plane lattice constant (3.35 Å). Overall calculated bands were contracted by 13 % in the energy axis to find a reasonable matching with the experiment. (c),(d) Experimental band structure near E_F , measured along the Γ M and Γ K cuts, respectively. Red and purple dashed curves are a guide for the eyes to trace the Te 5p and V 3d bands, respectively.

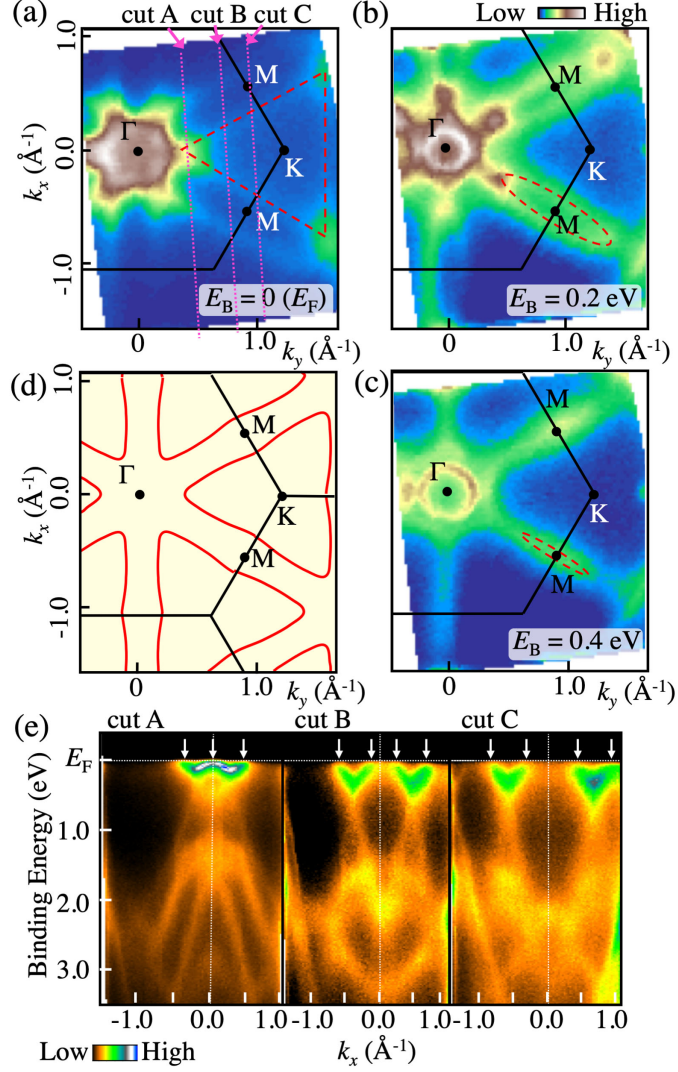


FIG. 3. (a)-(c) Plots of ARPES intensity at $T = 40$ K as a function of 2D wave vector, k_x and k_y , at three representative energy slices at $E_B = E_F$, 0.2 eV, and 0.4 eV, respectively. Energy contours were obtained by integrating the intensity within ± 50 meV with respect to each E_B 's. (d) Calculated FS obtained from the first-principles band-structure calculations for monolayer 1T-VTe₂. (e) ARPES-derived band structure along three representative $\mathbf{k}$ cuts [cuts A-C in (a)] which cross the triangular FS. The systematic evolution of the V-shaped band dispersions from cut A to cut C indicates the holelike nature of triangular FS.

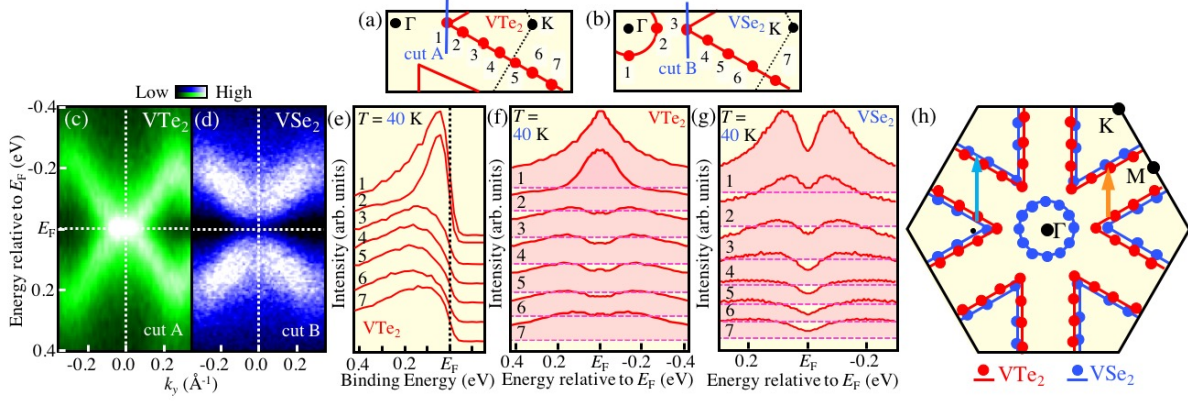


FIG. 4. (a),(b) Schematic FS together with a k cut and k points where high-resolution ARPES measurements were performed for monolayer VTe₂ and VSe₂, respectively. (c),(d) Plots of ARPES intensity as a function of wave vector and E_B symmetrized with respect to E_F , measured at $T = 40$ K along a cut crossing the corner of triangular pocket (cut A/B) for monolayer VTe₂ and VSe₂, respectively. (e) EDCs near E_F at $T = 40$ K for monolayer VTe₂ measured at various k_F points in (a). (f) Same as (e) but symmetrized with respect to E_F . Zero intensity for each spectrum is indicated by a dashed magenta line to highlight the absolute spectral weight. (g) Same as (f) but for monolayer VSe₂⁶. (h) Comparison of the FS topology between monolayer VTe₂ and VSe₂. Red and blue circles correspond to the k_F points for monolayer VTe₂ and VSe₂, respectively. Orange and light blue arrows indicate possible nesting vectors q for monolayer VTe₂ and VSe₂, respectively.