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μ-ARPES study of epitaxial atomic-layer VTe_2 film

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1 Introduction

Atomically thin transition-metal dichalcogenides (TMDs) are attracting particular attention because of a variety of the quantum physics emergent in atomically-thin limit, distinct from three-dimensional (3D) counterpart. VTe_2 is an excellent platform to address the evolution of physical properties upon the 2D-3D crossover, because the electronic property shows an intriguing difference between monolayer and bulk. Bulk VTe_2 forms the monoclinic 1*T*" phase with 3 × 1 × 3 periodic lattice distortion (PLD) and undergoes a structural transition to the polymorphic 1*T* structure at $T = 482$ K. On the other hand, monolayer VTe_2 stabilizes with the 1*T* structure and exhibits the 4 × 4 charge density wave (CDW). Besides the distinct phases in two extremal limits of VTe_2 (bulk and monolayer), another periodicity has been reported in multilayer VTe_2, but there is no consensus among research groups on physical properties such as CDW, crystal structure, and electronic states, triggering intensive debates on the origin of physical properties for multilayers. This problem apparently hinders from revealing the 2D-3D crossover in VTe_2. To clarify the evolution of electronic structure of VTe_2 from 2D to 3D, we have performed *in-situ* angle-resolved photoemission spectroscopy (ARPES) of monolayer and multilayer VTe_2 films grown by molecular-beam-epitaxy (MBE) method [1].

2 Experiment

Bilayer graphene was fabricated by direct resistive heating of a n-type Si-rich 4H-SiC(0001) wafer at 1100◦C at 1×10^{-9} Torr for 30 min. Then, VTe_2 film was grown by depositing V atoms on bilayer graphene in Te atmosphere. Number of layers are controlled by the deposition time while keeping the same deposition rate. The as-grown film was annealed at 300 ˚C for 30 min and then transferred to the μ-ARPES system [2] directly connected to the MBE system.

ARPES measurements were carried out using μ-ARPES system at the beamline BL- 28A in Photon Factory, KEK. Linear horizontal polarized light of 74 eV was used to excite photoelectrons. The energy and angular resolutions were set to be 20 meV and 0.2◦, respectively. The sample was kept at $T = 40$ K during the ARPES measurements.

3 Results and Discussion

Figures 1(a) and 1(b) show the low-energy-electron-diffraction (LEED) pattern of 1 and 6 monolayer (ML) VTe_2 films, respectively, grown on bilayer graphene/4H-SiC(0001). We clearly recognize in Fig. 1(a) a sharp 1 × 1 spot associated with the formation of 1 ML VTe_2 film, together with the graphene spots. On the other hand, when the film thickness is increased to 6 ML, the 2 × 2 spot (red arrow) emerges and the graphene spot disappears. This cannot be explained in terms of the formation of monoclinic 1*T*" phase seen in bulk VTe_2, because the periodicity of 1*T*" phase is 3 × 1.

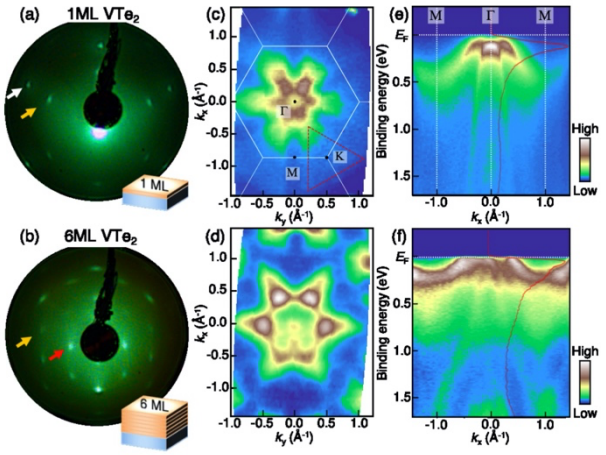

Fig. 1 (a) and (b) LEED pattern of 1 and 6 ML VTe_2, respectively. Orange, red and white arrows represent the spots/streaks from 1 × 1 VTe_2, 2 × 2 VTe_2, and bilayer graphene, respectively. (c) and (d) ARPES intensity plot at E_F of 1 and 6 ML VTe_2, respectively. (e) and (f) Band dispersion of 1 and 6 ML VTe_2 along the k_x cut measured with $h\nu = 74$ eV at $T = 40$K, respectively [1].

Figures 1(c) and 1(d) display the plot of ARPES intensity at E_F for 1 and 6 ML, respectively, measured at $T = 40$ K. In 1 ML, one can recognize a large triangular Fermi surface (FS) centered at the K point in the Brillouin zone (BZ) of 1*T* phase, with an enhanced intensity around the corner of triangular pocket near the Γ point. On the other

hand, the ARPES intensity for 6 ML displays a snowflake shape centered at Γ. Also, the intensity profile away from Γ is very different between 1 and 6 ML. Such a difference in the FS topology is also reflected in the experimental band dispersion shown in Figs. 1(e) and 1(f). Namely, the bottom of V 3*d* band is located at the binding energy (E_B) of ~0.5 eV at the M point for 1 ML, while it is pushed upward by ~0.3 eV for 6 ML, resulting in the narrower bandwidth along the k_x cut. Moreover, although the V 3*d* band for 1 ML does not touch E_F along the k_x cut [Fig. 1(e)], it apparently crosses E_F for 6 ML [Fig. 1(f)]. These results indicate that the electronic structure strongly depends on the number of VTe_2 layers.

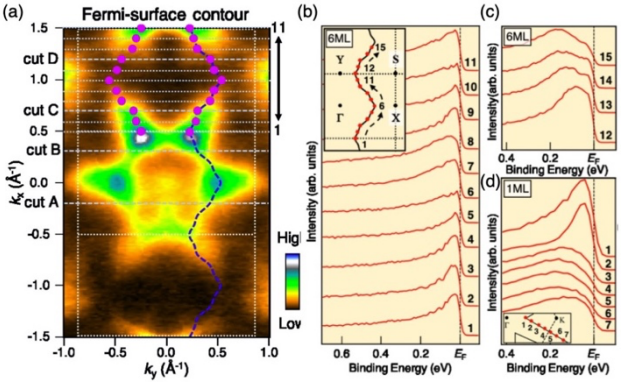

Fig. 2 (a) FS mapping of 6ML 1T'-VTe_2. Experimental Fermi wave vectors (k_F's; purple dots) are overlaid by dashed blue curve. (b) and (c) EDCs near E_F at T = 40 K for 6 ML VTe_2 measured at various k_F points on the quasi-1D FS [points 1–11 and 12–15 (see inset), respectively]. (d) EDCs at the representative k_F points on the triangular FS for 1 ML 1*T* -VTe_2 [1,3].

Figure 2(a) shows the FS mapping and experimental Fermi wave vectors (k_F's; purple dots) estimated from the momentum distribution curves (MDCs) at E_F. As shown in Fig. 2(a), the FS shows an open quasi-1D character with a strong warping effect as opposed to 1 ML with a simple 2D triangular pocket [Fig. 1(c)].

We show in Figs. 2(b) and 2(c) the energy distribution curve (EDC) at T = 40 K for 6 ML measured at various k_F points on the quasi-1D FS. In the first BZ of single-domain 1*T*' phase [points 1–11; Fig. 2(b)], one can see a peak around E_F followed by a sharp Fermi-edge cutoff. The Fermi edge is also recognized in the second BZ [points 12–15; Fig. 2(c)], whereas the EDC is complicated by the presence of another peak at E_B ~ 0.1–0.2 eV. These results suggest the absence of an energy gap on the entire quasi-1D FS at T = 40 K, in contrast to the 1 ML counterpart showing an anisotropic pseudogap highlighted in Fig. 2(d). We found from the analysis of reflection-high-energy-electron-diffraction (RHEED) patterns that the 1T' structure is immediately formed when the film thickness reaches 2 ML during epitaxy. The observed intriguing difference in the gap-opening behavior between single- and multilayer VTe_2 is explained in terms of the difference in their crystal phases (1T vs 1T') and the change in the FS topology and associated CDW properties. The present result demonstrates an important role of interlayer effect for controlling the crystal phase, and paves a pathway toward understanding the interplay between the fermiology, structural phases, and electronic properties in ultrathin TMDs [1].

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