Interfacial superconductivity induced by single-quintuple-layer Bi$_2$Te$_3$ on top of FeTe forming van-der-Waals heterostructure

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We report the first clear observation of interfacial superconductivity on top of FeTe(FT) covered by one quintuple-layer Bi$_2$Te$_3$(BT) forming van-der-Waals heterojunction. Both transport and scanning tunneling spectroscopy measurements confirm the occurrence of superconductivity at a transition temperature $T_c = 13$ K, when a single-quintuple-layer BT is deposited on the non-superconducting FT surface. The superconductivity gap decays exponentially with the thickness of BT, suggesting it occurs at the BT-FT interface and the proximity length is above 5 - 6 nm. We also measure the work function’s dependence on the thickness of BT, implying a charge transfer may occur at the BT/FT interface to introduce hole doping into the FT layer, which may serve as a possible candidate for the superconducting mechanism. Our BT/FT heterojunction provides a clean system to study the unconventional interfacial superconductivity.

The interfacial superconductivity has been of great interest $^{[1-7]}$, since it may help to resolve the mystery of high temperature superconductors with layered structures and to search for the topological superconductors $^{[8-10]}$. There are several mechanisms proposed for the interfacial superconductivity, including edge misfit dislocation defect $^{[2,11]}$, electric field gating $^{[3,4]}$ as well as chemical doping (e.g. charge transfer) $^{[5,6]}$. In order to pin down the superconducting mechanism unambiguously, the high quality sample with atomic abruptness at the interfaces is required. In the past decade, van der Waals (vdW) epitaxy turns out to be an effective way to grow two-dimensional interfacial superconductors $^{[8-10,12]}$, where the interface is usually of high quality even against the large lattice mismatch since the interfacial interaction at the heterojunctions is of vdW type.$^{[13]}$

Bi$_2$Te$_3$/Fe$_{1+x}$Te is one of the first realized van der Waals heterostructures which hosts an interfacial superconductivity between two non-superconductors Bi$_2$Te$_3$ and Fe$_{1+x}$Te $^{[8,9]}$, where Fe$_{1+x}$Te may have an unusual mechanism for superconductivity and magnetic order of ’11’ group iron based superconductors $^{[14,15]}$. A transport study showed that an optimal superconductivity ($T_c = 11.5$ K) only can be developed when the Bi$_2$Te$_3$ layer reaches a thickness of 5 quintuple-layer (QL) while single QL Bi$_2$Te$_3$/Fe$_{1+x}$Te only possesses a low $T_c$ around 2 K, indicating Bi$_2$Te$_3$ thickness may be crucial for the formation of superconductivity $^{[9]}$. In the past years, great efforts have been devoted to studying the intriguing superconductivity of Bi$_2$Te$_3$/Fe$_{1+x}$Te $^{[16,20]}$. For instance, Gu et al. reported a scanning tunneling microscopy (STM) study on the Bi$_2$Te$_3$ grown on Fe$_{1+x}$Te, however, the energy gap of superconductivity didn’t appear even Bi$_2$Te$_3$ is thicker than 6 QLs, instead a merging of Dirac electrons and the correlation effect was revealed in the scanning tunneling spectroscopy (STS), implying the thickness of Bi$_2$Te$_3$ is not the exclusive cause of superconductivity $^{[16]}$. In 2017, Manna et al. studied a reverse structure, Fe$_x$Te grown on bulk Bi$_2$Te$_3$ crystal, and probed an energy gap of superconductivity co-existing with bilinear antiferromagnetic (AFM) order $^{[18]}$, however, the gap revealed $T_c$ is lower than 6 K, inconsistent with the previous transport transport results $^{[9]}$, as in their sample the Bi$_2$Te$_3$ layer is thicker than the optimal 5 QLs. Obviously, the mechanism for Bi$_2$Te$_3$/Fe$_{1+x}$Te superconductivity is still a matter of debate, including the location of superconductivity and the possible driving factors, nevertheless, the revealed correlation effect and unusual AFM order made the system very attractive for studying the unusual mechanism for iron based superconductors.

In this work, we report the STM study of high quality Bi$_2$Te$_3$/FeTe interface grown on SrTiO$_3$(001) (STO) substrates. Remarkably, we found that an interfacial superconductivity immediately appeared once one QL Bi$_2$Te$_3$ patched the FeTe surface, with a high $T_c$ around 13 K confirmed by STS and transport study simultaneously. Furthermore, we present the first superconducting energy gap evolution of Bi$_2$Te$_3$/FeTe interface, implying that the superconductivity on the thicker Bi$_2$Te$_3$(2-5QL) layer is proximity-induced and the superconductivity is essentially located at the Bi$_2$Te$_3$-FeTe interface (includ-
FIG. 1. (a) A schematic illustration for 1 QL Bi$_2$Te$_3$ grown on FeTe. (b) A lattice comparison for a Bi$_2$Te$_3$/FeTe interface. (c) A HRSTEM image containing a Bi$_2$Te$_3$ (1 QL), a FeTe (20 nm) thin film and a STO substrate simultaneously. (d) A HRTEM image of a lateral heterojunction formed by a Bi$_2$Te$_3$ quintuple layer and a FeTe atomic step. (e) A lattice comparison for the lateral heterojunction and its nearby FeTe.

The samples were grown on single crystal 0.7% wt Nb-doped STO(001) substrates. The STO substrate was first gradually heated to about 950°C by electron-beam heating and annealed at this temperature for about 30 minutes. The substrate was then transferred in-situ to an adjacent Createc molecular beam epitaxy (MBE) system for sample growth. The FeTe thin films were grown in the MBE chamber by co-evaporation of high purity Fe (99.995%) and Te (99.999%) onto the substrate held at around 300 °C, with a Te rich flux ratio to ensure the stoichiometry. The thickness of FeTe layer is around 20 nm. The Bi$_2$Te$_3$ thin films were then grown by evaporating Bi$_2$Te$_3$ compound source at the substrate temperature of about 225 °C, with a growth rate around 2.5 Å/min for ensuring the precise QL control. Reflective high energy electron diffraction (RHEED) patterns (supplementary 1) kept streaky during the entire growth, indicating a layer-by-layer growth mode dominating the epitaxy. The samples were then transferred in-situ to a SPECS Joule-Thomson (JT) STM system. The base pressure in the MBE chamber and STO chamber was about 3 × 10$^{-10}$ mbar and 5 × 10$^{-10}$ mbar, respectively. The STM measurements were performed at 1.1 K (unless otherwise specified) with etched tungsten tips, which were sputtered with an Ar$^+$ ion sputter gun and tested on a reference Au(111). A high resolution scanning transmission electron microscopy (HRSTEM) was employed for a cross-sectional characterization on a Bi$_2$Te$_3$ (1 QL)/FeTe (20 nm) bilayer sample cut by a focus ion beam technique. Transport measurements were carried out with a standard four-point probe method using a commercial Quantum Design physical property measurement (PPMS) system. The ultraviolet photoemission spectroscopy (UPS) measurements were carried out using a SPECS PHOIBOS 150 hemispherical energy analyzer and a light source from a helium discharge lamp (He I, photon energy 21.218 eV).

Fig. 1 (a) suggests a cross-sectional schematic illustration of 1 QL Bi$_2$Te$_3$ grown on FeTe, well matching the lattice of the red square indicated region (Fig. 1 (b)) shown in the HRSTEM Fig. 1 (c), evidencing a sharp vDW interface without lattice distortion or inter-diffusion. Furthermore, a typical lateral Bi$_2$Te$_3$/FeTe heterojunction near a FeTe atomic step is imaged in Fig. 1 (d) and 1 (e), demonstrating a step-flow epitaxy. It is worth mentioning that no interstitial Fe atoms were detected by a careful examining the FeTe lattice shown in Fig. 1 (c). Combining with the 1:1 ratio of Fe/Te revealed by chemical analysis (see supplementary materials), we assert that the FeTe film is of strict stoichiometry, similar as previous reported stoichiometric FeTe thin films grown by MBE with Te rich flux [21].

The STM studies were carried out on more than 20 samples and the results are repeatable and consistent. Fig. 2 (a) is a typical topographic image after growth of about 1~2 QL of Bi$_2$Te$_3$, inside which we acquire atomic-resolution images of exposed FeTe surface and 1 QL Bi$_2$Te$_3$ surface shown in Fig. 2 (c) and (d), respec-
FIG. 2. (a) A STM topographic image of an area containing exposed FeTe, 1 QL and 2 QLs of Bi$_2$Te$_3$ (size: 220 × 220 nm$^2$, $V_{\text{Bias}} = 2.0$ V, $I_{\text{Tunnel}} = 50$ pA, color scale: 1.57 nm). (b) A STM topographic image showing how Bi$_2$Te$_3$ typically grows on FeTe films (size: 100 × 100 nm$^2$, $V_{\text{Bias}} = 1.8$ V, $I_{\text{Tunnel}} = 50$ pA, color scale: 1.24 nm). (c) An atomic resolution image of the exposed FeTe surface (size: 8 × 8 nm$^2$, $V_{\text{Bias}} = -10$ mV, $I_{\text{Tunnel}} = 200$ pA, color scale: 0.03 nm). (d) An atomic resolution image of the 1 QL Bi$_2$Te$_3$ surface (size: 8 × 8 nm$^2$, $V_{\text{Bias}} = 6.5$ mV, $I_{\text{Tunnel}} = 400$ pA, color scale: 0.03 nm). (e) A line profile corresponding to the blue line in (b). (f) Representative $dI/dV$ spectra acquired on the FeTe surface, 1 QL Bi$_2$Te$_3$, and 2 QL Bi$_2$Te$_3$ terraces, respectively (set point: $V_{\text{Bias}} = 10$ mV, $I_{\text{Tunnel}} = 50$ pA). (g) Representative $dI/dV$ spectra in a wider bias range acquired on the FeTe, 1 QL Bi$_2$Te$_3$, and 2 QL Bi$_2$Te$_3$ terraces, respectively (set point: $V_{\text{Bias}} = 0.3$ V, $I_{\text{Tunnel}} = 50$ pA). (h) Temperature dependent resistance $R$-$T$ curve of a sample of 1nm Bi$_2$Te$_3$/40 nm FeTe/STO under different magnetic fields perpendicular to the interface. The inset in (h) shows the $R$-$T$ curve of the same sample in a wider temperature range without applying magnetic field. (i) Temperature dependent $dI/dV$ spectra acquired on the 1 QL Bi$_2$Te$_3$ (set point: $V_{\text{Bias}} = 10$mV, $I_{\text{Tunnel}} = 50pA$). The texts in the all the STM images indicate the respective areas identified. Measured temperature is 1.1 K.

respectively, representing a perfect lattice without any adatoms and vacancies being observed. The growth time for the sample in Fig. 2(b) was extremely short to observe the growth behavior, showing a nucleation of Bi$_2$Te$_3$ takes place on the atomic step of FeTe, consistent with cross-sectional HRSTEM results shown in Fig. 1. Fig. 2(e) is the line profile corresponding to the blue line in Fig. 2(b). The step height of about 0.6 nm and 1.0 nm correspond to one unit cell of FeTe and 1 QL layer of Bi$_2$Te$_3$, respectively. Representative $dI/dV$ spectra in a narrow bias range acquired on the FeTe, 1 QL Bi$_2$Te$_3$, and 2 QL Bi$_2$Te$_3$ terraces are shown in Fig. 2(f). $dI/dV$ measures the local DOS near the Fermi level. The spectra acquired on the 1 QL Bi$_2$Te$_3$ terrace show two clear coherent superconducting peaks around 2.5 meV. This is in clear contrast to the spectrum acquired on the FeTe surface, which shows no coherent peaks, but a valley shape spectrum with a flat bottom. On the 2 QL Bi$_2$Te$_3$, the spectrum also exhibits two coherent superconducting peaks; however, the peaks are smaller at around 1.8 meV. The spectra on each terrace also vary little, indicating a good uniformity of the superconductivity. The temperature dependent $dI/dV$ spectra on the 1 QL Bi$_2$Te$_3$ surface are shown in Fig. 2(i), while the results on the 2 QL are in the Supplementary Materials [22]. While no significant change in the intensity of the coherence peak for the spectra is observed for temperature below 6 K, there is a significant decrease in the peak intensity at temperatures above 6 K. The gap magnitude is discernible up to about 10 K. At 12 K, there is no clear gap observed, consistent with followed transport revealed $T_c$. Typical $dI/dV$ spectra in a wider bias range obtained in the FeTe, 1 QL Bi$_2$Te$_3$, and 2 QL Bi$_2$Te$_3$ surfaces are plotted in Fig. 2(g). According to the $dI/dV$ spectrum Bi$_2$Te$_3$ film
of 1 QL, there is discernibly weak and nearly constant density of states from 0 eV (Fermi level) to -0.43 eV. Below about -0.43 eV, the density of states start to rise rapidly with a kink at about -0.55 eV. On the Bi$_2$Te$_3$ surfaces, respectively. Spectra were shifted vertically for clarity. (b) Gap magnitude versus the Bi$_2$Te$_3$ thickness.

The temperature dependent in-plane resistance ($R$-$T$ curve) of a Bi$_2$Te$_3$/FeTe/STO device with an average Bi$_2$Te$_3$ thickness of around 1 QL is shown in Fig. 2 (h). In the zero field (the inset in Fig. 2 (h)), a broad transition at about 65 K is observed, likely related to the magnetic and/or structural transition of FeTe films [14, 24-29]. The $R$-$T$ curve shows a superconducting transition with an onset temperature of $T_c^{\text{onset}} = 13$ K and zero resistance temperature of $T_c = 7.5$ K. Before $T_c^{\text{onset}}$, there is a upturn in $R$-$T$ curve with decreasing temperature. The magnetic field (applied perpendicular to the sample interface) suppresses the superconductivity, and 12 T field is far away to completely kill the superconductivity, indicating a very high critical field, which is a characteristic feature of FeTe based superconductors [30].

The $dI/dV$ spectra were acquired on samples with thicker Bi$_2$Te$_3$ films up to about 5-6 QL and are all shown in Fig. 3 (a). As can be seen, all of these spectra exhibit two coherent peaks, whereas the gap magnitude decreases with increasing Bi$_2$Te$_3$ thickness. The gap magnitude $\Delta$ of the spectra is plotted in Fig. 3 (b) with respect to the number of QLs of Bi$_2$Te$_3$. The $\Delta$ shows an exponential decay with a relationship of $\Delta = 3 \times 10^{-2} eV$ (in unit of meV, $N$ is the number of QLs of Bi$_2$Te$_3$) as obtained from the fitting, consistent with the characteristic of proximity-induced superconductivity [12], from which we can infer that the superconductivity is not enhanced in any way when the Bi$_2$Te$_3$ is 2 QLs or more than that when the Bi$_2$Te$_3$ is only 1 QL; instead, the superconductivity locates in the 1 QL Bi$_2$Te$_3$/FeTe interface, and becomes weaker on the Bi$_2$Te$_3$ surface due to the proximity effect.

To further unveil the charge transfer mechanism between Bi$_2$Te$_3$ and FeTe, we also in-situ measured the work function of the FeTe film before and after the growing of Bi$_2$Te$_3$ film with average thickness of 0.5 QL, 2 QL, 3 QL, and 6 QL, respectively using UPS at 15 K. The UPS result of FeTe film is shown in Fig. 4 (a). A voltage $-2$ V was applied between the sample and the spectrometer so that the photoelectrons were accelerated and then the low energy inelastic electrons could be distinguished from secondary electrons generated in the spectrometer by impact [31], which can be seen in Fig. 4 (a) as a small peak near 5.5 eV. The work function is calculated with the formula $\Phi = h\nu - E_{\text{in}}$, where $h\nu$ is the photon energy and $E_{\text{in}}$ is the spectrum width, the distance between the low energy cutoff and the Fermi edge as shown in Fig. 4 (a). Then we measured the photoemission spectra of Bi$_2$Te$_3$ covered- FeTe with increasing thickness.
of Bi$_2$Te$_3$ and the results are shown in Fig. 3 (b). The zoom-in view of the low kinetic energy region of Fig. 3 (b) is shown in Fig. 3 (c), depicting that the inelastic edge offsets to the high kinetic energy range as the thickness of Bi$_2$Te$_3$ being increased, while the fermi edge in the high energy end is pinned in all the spectrums as they are determined by the accelerating voltage (~3 V) solely. The extracted work functions are shown in Fig. 3 (d). As can be seen, the work function of FeTe film is about 4.80 eV. With the growth of Bi$_2$Te$_3$, the work function increases gradually, and starts to saturate when the Bi$_2$Te$_3$ thickness reaches about 3 QL. This result clearly suggests that there is a doping effect when the Bi$_2$Te$_3$ film is deposited onto the FeTe film, i.e., FeTe is hole-doped and Bi$_2$Te$_3$ film is electron-doped, qualitatively consistent the STS revealed charge transfer in Fig. 3 (g).

In the heterostructure of Bi$_2$Te$_3$/FeTe in our study, neither Bi$_2$Te$_3$ or FeTe is superconducting. Upon doping, Bi$_2$Te$_3$ is only superconducting with a maximum reported transition temperature of 5.5 K [22]. On the other hand, FeTe is superconducting upon doping with either oxygen [30] or selenium [33] with a transition temperature of about 14 K, which is surprisingly close to the transition temperature in our study. Our work function measurements clearly show that there is a hole doping effect on the FeTe layer in the heterostructure. Therefore, it is plausible that the single FeTe layer become superconductivity due to the loss of electrons induced by Bi$_2$Te$_3$ covering, leading to the interface superconductivity observed in our study. While it still remains a challenge on how this doping leads to the observed superconductivity, our results provide more insights for future theoretic work to understand this doping induced change on electronic and magnetic properties of a single FeTe layer, which may also shed light on resolving the mystery of iron based superconductivity.

In conclusion, the superconductivity of Bi$_2$Te$_3$/FeTe system is studied by using the low temperature scanning tunneling microscopy. It is found that the superconductivity in the Bi$_2$Te$_3$/FeTe system can be induced by only one QL of Bi$_2$Te$_3$, which is also confirmed by the transport measurements. Scanning tunneling spectroscopy further shows that the superconducting gap decays exponentially with the increasing Bi$_2$Te$_3$ thickness, implying the superconductivity in Bi$_2$Te$_3$ bulk is actually proximity-induced by the interface. Our results provide unambiguous evidence that the superconductivity in the Bi$_2$Te$_3$/FeTe system is located around the Bi$_2$Te$_3$/FeTe interface. As for the superconducting mechanism, it is found that the doping of FeTe may be possible to generate superconductivity in the Bi$_2$Te$_3$/FeTe vdW heterojunctions.

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