Studies on the origin of the interfacial superconductivity of $\text{Sb}_2\text{Te}_3/\text{Fe}_{1+y}\text{Te}$ heterostructures

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The recent discovery of the interfacial superconductivity (SC) of the $\text{Bi}_2\text{Te}_3/\text{Fe}_{1+y}\text{Te}$ heterostructure has attracted extensive studies due to its potential as a novel platform for trapping and controlling Majorana fermions. Here we present studies of another topological insulator (TI)/ Fe_{1+y}Te heterostructure, $\text{Sb}_2\text{Te}_3/\text{Fe}_{1+y}\text{Te}$, which also has an interfacial 2-dimensional SC. The results of transport measurements support that reduction of the excess Fe concentration of the Fe_{1+y}Te layer not only increases the fluctuation of its antiferromagnetic (AFM) order but also enhances the quality of the SC of this heterostructure system. On the other hand, the interfacial SC of this heterostructure was found to have a wider-ranging TI-layer thickness dependence than that of the $\text{Bi}_2\text{Te}_3/\text{Fe}_{1+y}\text{Te}$ heterostructure, which is believed to be attributed to the much higher bulk conductivity of Sb_2Te_3 that enhances indirect coupling between its top and bottom topological surface states (TSSs). Our results provide evidence of the interplay among the AFM order, itinerant carriers from the TSSs, and the induced interfacial SC of the TI/ Fe_{1+y}Te heterostructure system.

iron-based parent compound | topological insulator | interfacial superconductivity | spin fluctuation of antiferromagnetic order | itinerant carriers of topological surface states

Three-dimensional topological insulators (3D TIs) with extraordinary electronic states have attracted intense research interest in recent years due to their unusual electronic band structures (1–4). In an ideal 3D TI, bulk states feature an energy gap like an ordinary insulator, while surface states are characterized by a linear Dirac-like dispersion energy band with spin texture locked helically to momentum, resulting in metallic topological surface states (TSSs). The most studied 3D TIs include Bi_2Se_3 , Bi_2Te_3 , and Sb_2Te_3 binary compounds that have a rhombohedral structure in which the unit cell consists of 3 quintuple layers (QL) bonded by van der Waals (vdW) force. Heterostructures containing a 3D TI are extensively studied because they are expected to produce novel phenomena that may not arise from heterostructures composed of conventional materials. For example, topological superconductivity (SC) has been theoretically predicted to be induced on the surface of TI-superconductor heterostructure via a proximity effect, which may provide a platform to host non-Abelian Majorana fermions (5, 6). Interestingly, our group recently discovered a 2-dimensional (2D) SC at the interface of 2 nonsuperconducting materials (7, 8), Fe_{1+y}Te , a parent compound of iron-based superconductors, and Bi_2Te_3 , a 3D TI, and later we reported further studies on this heterostructure such as the superconducting proximity effect (9), current-induced depairing (10), anisotropic magnetic responses (11), and vortex dynamics (12, 13). Due to the complexity of this heterostructure, revealing the underlying mechanism of its induced 2D SC with a theoretical approach is very unlikely. Recently, experimental studies using angle-resolved photoemission spectroscopy (ARPES) (14) and ultraviolet photoemission spectroscopy (15) reported that the Fe_{1+y}Te layer in this heterostructure involved hole doping through

a transfer of electrons into the Bi_2Te_3 layer. However, confirmation of this claim requires further studies to provide evidence about the linkage between the charge transfer and the behavior of the observed 2D SC.

The 2D SC of the $\text{Bi}_2\text{Te}_3/\text{Fe}_{1+y}\text{Te}$ heterostructure is likely induced on the Fe_{1+y}Te layer, as described in several previous works (9, 15). The studies on how some physical parameters involved in this heterostructure could modify the physical property of the Fe_{1+y}Te layer and its 2D SC may provide hints to achieve a better understanding of the underlying mechanism of the induced SC. One of the most interesting issues in iron-based superconductors is the interplay between magnetism and SC. A synthesized bulk Fe_{1+y}Te crystal usually favors excess Fe atoms in interstitial sites, which could affect its magnetic order and electronic topological Lifshitz transitions (16) as in doped iron-based superconductors (17). Koz et al. (18) previously reported that below 70 K, for bulk Fe_{1+y}Te with $0.04 < y < 0.11$, the spin direction of Fe atoms is oriented along the diagonal of the Fe–Fe square lattice, forming a bicollinear commensurate antiferromagnetic (AFM) structure, followed by a mixed phase for $0.11 < y < 0.13$ and then an incommensurate AFM magnetic order for $0.13 < y < 0.15$ as y increases. They also observed that the commensurate AFM transition temperature for bulk Fe_{1+y}Te increases as the concentration of excess Fe decreases down to $y = 0.04$. Studies on superconducting iron

Significance

Interfacial superconductivity (SC) is found in the $\text{Sb}_2\text{Te}_3/\text{FeTe}$ heterostructure system, of which neither the Sb_2Te_3 topological insulator (TI) nor the FeTe parent compound of iron chalcogenides is superconducting. We have systematically studied some factors that are responsible for inducing the 2-dimensional SC, including the spin fluctuation of antiferromagnetic ordering in Fe_{1+y}Te layer and the thickness of Sb_2Te_3 . These findings help us understand the mechanism of the unconventional SC at the interface of TI/FeTe heterostructures. TI/FeTe heterostructures will provide a new platform for trapping and controlling Majorana fermions.

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chalcogenide ternary $\text{Fe}_{1.02}\text{Te}_{1-x}\text{Se}_x$ (19) and iron pnictides such as $\text{ReFeAsO}_{1-x}\text{F}_x$ (Re = rare earth element) (20) showed that AFM order is antagonistic to SC that generally occurs after the magnetic order was suppressed by varying chemical doping concentration, even though they can coexist in a certain composition range (21, 22).

In this study, we demonstrate that 2D SC also appears in another TI/ Fe_{1+y}Te system, the $\text{Sb}_2\text{Te}_3/\text{Fe}_{1+y}\text{Te}$ heterostructure. We have systematically studied how the quality of the 2D SC of this heterostructure depends on the fluctuation of the AFM order of the Fe_{1+y}Te layer and the thickness of the Sb_2Te_3 layer. Particularly, the results from the former study provide a deeper understanding on the correlation between SC and AFM in this heterostructure, while the results from the latter study support that the TSSs may be involved in the induction of SC. Based on these experimental observations, we propose a possible origin of the induced interfacial SC.

Results

Structural Characterizations of the $\text{Sb}_2\text{Te}_3/\text{Fe}_{1+y}\text{Te}$ Heterostructure. Reflection high-energy electron diffraction (RHEED) patterns were captured after the growth of Fe_{1+y}Te and Sb_2Te_3 layers. Fig. 1 *A* and *B* display the RHEED patterns of tetragonal Fe_{1+y}Te when the incident electron beam is along the Fe_{1+y}Te [100] and $[1\bar{1}0]$ axis with the sample rotation angle at $\varphi = 0^\circ$ and $\varphi = 45^\circ$, respectively. The diffraction streaks are marked by short lines and denoted by 2D Miller indices. The streaky patterns of Fe_{1+y}Te display a 90° rotational symmetry, as expected from the

tetragonal structure of Fe_{1+y}Te . In contrast, the RHEED pattern of Sb_2Te_3 at rotation angle $\varphi = 0^\circ$ shown in Fig. 1*C* has 2 sets of streaks. The spacing ratio of the wider and narrower set of streaks is approximately $\sqrt{3}$, and their 2D Miller indices are marked correspondingly. As the sample rotates, the diffraction pattern of Sb_2Te_3 repeats at every 30° , which can be explained by the fact that the as-grown Sb_2Te_3 layer consists of 2 hexagonal lattices twisted by 90° . This is attributed to the 4-fold symmetry of the bottom Fe_{1+y}Te lattice and the 6-fold symmetry of the top Sb_2Te_3 lattice with respect to the growth direction. The details will be addressed in the next paragraph where the results of cross-sectional scanning transmission electron microscopy (STEM) imaging are presented.

The epitaxial growth of bismuth and antimony chalcogenides on substrates that have 6-fold symmetry, such as $\text{Al}_2\text{O}_3(0001)$ (23), $\text{Si}(111)$ (24), $\text{BaF}_2(111)$ (25, 26), $\text{InP}(111)$ (27), and $\text{GaAs}(111)$ (28), usually results in 2 mirror-symmetric twin domains due to the fact that there are 2 stacking sequences (ABCAB and ACBAC) along the [0001] direction for one QL of these chalcogenides. In order to investigate whether such twin domains exist in Sb_2Te_3 grown on Fe_{1+y}Te that has 4-fold symmetry and how they are developed into a different symmetry observed by RHEED as described above, high-resolution spherical-aberration-corrected STEM imaging was performed on an $\text{Sb}_2\text{Te}_3(27 \text{ QL})/\text{Fe}_{1+y}\text{Te}(60 \text{ nm})$ heterostructure (one QL of Sb_2Te_3 is approximately equal to $10.2\text{\AA}$), which was expected to provide more visualized and finer structural analysis at atomic scale than what can

Fig. 1. Structural analysis of a $\text{Sb}_2\text{Te}_3/\text{Fe}_{1+y}\text{Te}$ heterostructure. RHEED patterns of Fe_{1+y}Te taken when the electron beam is along the (A) [100] and (B) $[1\bar{1}0]$ direction. (C) RHEED pattern of Sb_2Te_3 taken when the electron beam is along the [100] direction of Fe_{1+y}Te . The short lines in A–C denote 2D Miller indices of the observed diffraction streaks. (D–F) Three cross-sectional STEM-HAADF images of the heterostructure taken at different locations of one TEM specimen with the zone axis along the [100] direction of Fe_{1+y}Te . (G and H) Schematic drawings of top and side views of the atomic arrangement of domains D and E, respectively. (I and J) Schematic drawings of top and side views of the other 2 domains that have identical side-view atomic arrangement corresponding to the STEM-HAADF image shown in F. The red and blue arrows marked in G–J indicate the crystalline axes [1010] and [0110] of Sb_2Te_3 , respectively. They are conventionally written as a and b axes for a hexagonal lattice.

be achieved by RHEED. Cross-sectional high-angle annular dark field (HAADF) STEM images of this heterostructure observed at different locations of one TEM specimen with the zone axis along the [100] direction of Fe_{1+y}Te are displayed in Fig. 1 D–F. As can be seen, Fig. 1 D and E display the atomic arrangement of Te-Sb-Te-Sb-Te QL stacking in ABCAB and ACBCB sequence with the [1120] direction (Fig. 1D) and $[\bar{1}\bar{1}20]$ direction of Sb_2Te_3 (Fig. 1E) aligned with the [100] direction of Fe_{1+y}Te , respectively. In Fig. 1 G and H, schematic drawings with both top-view and side-view modes for these 2 stacking sequences are displayed. As mentioned earlier, the Sb_2Te_3 layer was grown on $\text{Fe}_{1+y}\text{Te}(001)$ that has a 4-fold symmetry, and thus it is expected that one can find 2 other domains that are laterally twisted by 90° from each of the 2 domains schematically drawn in Fig. 1 G and H. Fig. 1 I and J show the corresponding drawings of these expected domains. One can see that these 2 domains have identical side-view atomic arrangement when the viewing zone axis is along the [100] direction of Fe_{1+y}Te . In fact, we did observe such an atomic arrangement among our STEM images, and one typical example is shown in Fig. 1F. These observations indeed provide evidence that the Sb_2Te_3 layer consists of 2 hexagonal lattices twisted by 90° . It is worth pointing out that the 4 different domains mentioned above can be generated through azimuthal rotation of a domain having a stacking sequence either shown in Fig. 1 G and H, then laterally twisted by 90° , 180° , and 270° . This can be visualized by starting with the domain in Fig. 1G, then Fig. 1 I, H, and J are the resulting domains when it is twisted by 90° , 180° , and 270° , respectively. It is also worth mentioning that all of the STEM-HAADF images shown in Fig. 1 D–F enjoy a sharp and defect-free interface, which can be attributed to the vdW bonding nature between Sb_2Te_3 and Fe_{1+y}Te . Based on these images, the lattice parameters of the Fe_{1+y}Te unit cell at the interface are determined to be $a = 3.88\text{\AA}$ and $c = 6.24\text{\AA}$, and those of Sb_2Te_3 are $a = 4.24\text{\AA}$ and $c = 30.54\text{\AA}$, which are close to their reported bulk values. High-resolution X-ray diffraction studies (SI Appendix, Fig. S1) support that all of the 4 domains of Sb_2Te_3 likely orient with their c -axes along the growth direction (29).

Two-Dimensional SC. Fig. 2 A and B show the temperature-dependent resistance of the $\text{Sb}_2\text{Te}_3(27\text{ QL})/\text{Fe}_{1+y}\text{Te}$ heterostructure measured under different magnetic fields in directions perpendicular (H) and parallel ($H_{\parallel}$) to the interface, respectively (29). At zero applied magnetic field, resistance starts to drop at an onset temperature T_C^{onset} of 12.3 K and zero-resistance state happens at 3.1 K. Fig. 2 A and B display a large anisotropy regarding the direction of the applied magnetic field since the transitions significantly broaden as $H_{\perp}$ increases, while such a broadening is much weaker as $H_{\parallel}$ increases to the same magnitudes, indicating the SC is likely in 2D. Its further confirmation could be obtained by studying the temperature dependence of the upper critical magnetic field $\mu_0 H_{c2}$ of the observed SC. Here, we define the critical superconducting transition temperature T_C as the resistance drops to 50% of the normal-state value at 15 K. The out-of-plane upper critical magnetic field and in-plane upper critical field are denoted as $\mu_0 H_{c2}^{\perp}$ and $\mu_0 H_{c2}^{\parallel}$, respectively. Fig. 2C shows the temperature dependence of the upper critical field $\mu_0 H_{c2}(T)$ for the $\text{Sb}_2\text{Te}_3(27\text{ QL})/\text{Fe}_{1+y}\text{Te}$ heterostructure in directions parallel ($H_{\parallel}$) and perpendicular ($H_{\perp}$) to the interface (29), where the x axis denotes the T_C values at different applied magnetic field. It is well known that 2D SC is governed by Ginzburg–Landau (GL) theory (30) with the temperature dependences of the upper critical magnetic fields expressed by

$$\mu_0 H_{c2}^{\perp} = \frac{\Phi_0}{2\pi\xi_{GL}^2(0)} \left(1 - \frac{T}{T_C(0)}\right) \quad [1]$$

$$\mu_0 H_{c2}^{\parallel} = \frac{\Phi_0\sqrt{12}}{2\pi\xi_{GL}(0)d_{SC}} \left(1 - \frac{T}{T_C(0)}\right)^{\frac{1}{2}}, \quad [2]$$

where Φ_0 is the magnetic flux quantum, $\xi_{GL}(0)$ the zero-temperature GL in-plane coherence length, d_{SC} the temperature-independent SC thickness, and $T_C(0)$ the critical temperature at zero magnetic

Fig. 2. Two-dimensional SC nature of the $\text{Sb}_2\text{Te}_3(27\text{QL})/\text{Fe}_{1+y}\text{Te}$ heterostructure. Resistance as a function of temperature under (A) out-of-plane and (B) in-plane magnetic field up to 12 T. (C) Temperature dependence of the upper critical field $\mu_0 H_{c2}^{\perp}$ and $\mu_0 H_{c2}^{\parallel}$. Solid curves are obtained by fitting with the theoretical 2D GL equations. (D) Temperature dependence of resistance with a fitted curve based on the BKT model, yielding $T_{\text{BKT}} = 3.68\text{ K}$. (Inset) $[d(\ln R/dT)]^{-2/3}$ as a function of temperature with its linear part extrapolated to zero, yielding $T_{\text{BKT}} = 3.64\text{ K}$. (E) V - I curves at various temperatures plotted on a logarithmic scale. The line with $\alpha = 1$ represents ohmic behavior while the line with $\alpha = 3$ corresponds to the BKT transition. (F) α versus T extracted from fitting the data in E with power law $V \propto I^\alpha$, showing α approaches 3 at $T_{\text{BKT}} = 3.71\text{ K}$.

field. Our data shown in Fig. 2C indeed can be well fitted by this theory in both parallel and perpendicular directions; the solid curves in this figure result from the best fitting with $\xi_{GL}(0) = 4.1 \pm 0.2$ nm and $d_{SC} = 3.9 \pm 0.2$ nm. It is important to point out that neither a pure Fe_{1+y}Te nor a pure Sb_2Te_3 thin film grown under similar conditions exhibits SC (SI Appendix, Fig. S2), further supporting that the observed SC in the $\text{Sb}_2\text{Te}_3/\text{Fe}_{1+y}\text{Te}$ heterostructure occurs at its interface.

It is commonly accepted that the transport properties of a 2D SC system can be well described by the 2D Berezinskii–Kosterlitz–Thouless (BKT) theoretical model (31–33), in which vortex–antivortex pairs are formed and bound together below a critical temperature called the BKT temperature, T_{BKT} . At a temperature above T_{BKT} , vortex–antivortex pairs are thermally dissociated and free vortices give rise to a finite resistance. At a temperature just above T_{BKT} , the temperature-dependent resistance at zero magnetic field is predicted to be in the form of

$$R = R_0 \exp(-bt^{-1/2}), \quad [3]$$

where R_0 and b are material-specific parameters and $t = (T/T_{BKT}) - 1$ is the reduced temperature. The main panel of Fig. 2D shows the best fit using Eq. 3 for the temperature-dependent resistance at zero applied magnetic field of the heterostructure, yielding $T_{BKT} = 3.68$ K. Fig. 2D, Inset shows the extrapolation of the linear part of $(\ln R/dT)^{-2/3}$ to zero, yielding a consistent value of $T_{BKT} = 3.64$ K (29). Another important piece of evidence of BKT transition is obtained from fitting the voltage–current (V – I) curves on a log–log scale for a temperature range from 2 to 12 K near each critical current in its superconducting transition region with a $V \propto I^{\alpha(T)}$ power-law dependence, where α equals 3 at T_{BKT} , as shown in Fig. 2E (29). Fig. 2F plots α as a function of temperature from which T_{BKT} is determined to be 3.71 K (29). The above analyses thus provide strong evidence for the 2D nature of the observed SC of the $\text{Sb}_2\text{Te}_3/\text{Fe}_{1+y}\text{Te}$ heterostructure.

The Role of Fluctuation of the AFM Order. Aiming at studying the interplay between the AFM order and SC, a group of $\text{Sb}_2\text{Te}_3(24 \text{ QL})/\text{Fe}_{1+y}\text{Te}(60 \text{ nm})$ heterostructures named SF-1, -2, and -3 and 2 pure $\text{Fe}_{1+y}\text{Te}(60 \text{ nm})$ thin films named F-1 and -2 were fabricated with various excess Fe concentrations. A combination of X-ray photoelectron spectroscopy (XPS) and spherical-aberration-corrected STEM was used to estimate the nominal y values of these samples. To achieve a more accurate analysis, a standard sample with known chemical composition is needed to derive the more reliable values of the relative sensitivity factors (RSFs) used in XPS analysis. These RSFs are further assumed to be constants for a certain variation of the sample matrix. We used sample SF-1 as such a standard sample and its y value was estimated by counting the number of interstitial Fe atoms in its spherical-aberration-corrected STEM images of its Fe_{1+y}Te layer. Fig. 3 shows one of the STEM-HAADF images that have the highest detected amount of interstitial Fe, in which the rectangular boxes contain the interstitial Fe atoms. Since fewer interstitial Fe atoms are seen in some of these images, we obtained the estimated y value of this sample as 0.004 by taking the average. The nominal y values of SF-2, SF-3, F-1, and F-2, as estimated using the peak areas of Fe and Te in their XPS spectra and the RSFs derived from the estimated y value of sample SF-1, are 0.021, 0.074, 0.042, and 0.09, respectively.

The normalized resistances as a function of temperature for samples F-1 and -2 are shown in Fig. 4A (29). Sample F-1 with fewer excess Fe concentration of the 2 samples shows a semiconducting behavior within the entire measured temperature range from 300 K to 2 K. Sample F-2 with more excess Fe shows a semiconducting-to-metallic transition at 47.3 K, which is known to be associated with the AFM phase transition. The spin fluctuation

Fig. 3. A cross-sectional STEM image of sample SF-1 taken in the Fe_{1+y}Te region. The red rectangles enclose the areas that have interstitial Fe atoms.

of the AFM order in Fe SCs was previously proposed to be correlated to their SC (34, 35). We believe that such fluctuation can be reflected by the distinctness of the AFM transition, that is, a less distinctive transition corresponds to higher spin fluctuation, which is consistent with the observations on the temperature-dependent resistivity curves of $\text{Fe}_{1.02}(\text{Te}_{1-x}\text{Se}_x)$ alloys (19). Interstitial Fe atoms are believed to provide magnetic proximity coupling with the neighboring AFM spins [as we addressed in a previous study (9)] to reduce their fluctuation. The results shown in Fig. 4A indicate that the reduction of excess Fe concentration in Fe_{1+y}Te layer increases the fluctuation of this magnetic ordering, consistent with the previous findings from Bao et al. (35) using a spin polarized inelastic neutron spectrometer. Fig. 4B shows the temperature-dependent normalized resistances of samples SF-1, -2, and -3 (29). Samples SF-1 and SF-2 with less excess Fe than sample SF-3 show a semiconducting behavior as the temperature decreases from 300 K to just above the superconducting transition temperature, while SF-3 shows an AFM transition at 47.6 K. This correlation is consistent with that observed for the 2 pure Fe_{1+y}Te layers as shown in Fig. 4A. As can be seen in Fig. 4B, the onset temperatures of superconducting transition are 9.2 K, 8.9 K, and 8.7 K for SF-1, -2, and -3, respectively. These results indicate that the reduction of excess Fe concentration not only increases the spin fluctuation of the AFM order in the Fe_{1+y}Te layer but also enhances the quality of the SC of this heterostructure system. In fact, this provides important evidence that spin fluctuation of the AFM order in the Fe_{1+y}Te layer of the heterostructure is related to the induction of the observed 2D SC.

Sb_2Te_3 Thickness Dependence on the Interfacial SC. In the following, we will present the studies on the Sb_2Te_3 layer thickness dependence of the 2D SC of the $\text{Sb}_2\text{Te}_3/\text{Fe}_{1+y}\text{Te}$ heterostructures with the aim of providing evidence that the TSSs of the Sb_2Te_3 layer are likely involved in the induction of the SC. A set of $\text{Sb}_2\text{Te}_3/\text{Fe}_{1+y}\text{Te}(60 \text{ nm})$ heterostructures, each containing an Sb_2Te_3 layer with a different thickness, was fabricated for these studies. Fig. 5A shows the normalized temperature-dependent resistance $R/R(T = 100 \text{ K})$ curves of $\text{Sb}_2\text{Te}_3/\text{Fe}_{1+y}\text{Te}$ heterostructures with Sb_2Te_3 of 10, 15, 24, and 27 QL, which display SC with onset temperatures of 3.7, 6.6, 8.7, and 12.3 K, respectively, showing a trend that the onset temperature of SC increases with the Sb_2Te_3 thickness (29). To better illustrate the SC signature of

Fig. 4. The interplay among the excess Fe concentration, magnetism, and SC. Normalized temperature-dependent resistance of (A) 2 pure $Fe_{1+y}Te$ (60-nm) films named F-1 and F-2 and (B) a group of Sb_2Te_3 (24 QL)/ $Fe_{1+y}Te$ (60-nm) heterostructures named SF-1, SF-2, and SF-3 with various excess Fe concentration y . (B, Inset) A zoomed-in region at low temperature from 1 K to 15 K for displaying the contrast of the SC transition of these heterostructures.

the $Sb_2Te_3(10 \text{ QL})/Fe_{1+y}Te$ sample with the thinnest Sb_2Te_3 layer among the 4 heterostructures, Fig. 5A, Inset shows the resistance versus temperature curve for this sample near the SC transition, which indeed displays a resistance drop at 3.7 K. Fig. 5B shows the zoomed-in normalized resistance curves for these heterostructure samples around their AFM transition temperatures, which are 48.2, 53.3, 47.6, and 53.3 K for Sb_2Te_3 thickness of 10, 15, 24, and 27 QL, respectively. The variation of the AFM transition temperatures of these heterostructures is caused by the small differences in the spin fluctuation of the AFM order of their $Fe_{1+y}Te$ layers. As can be seen in Fig. 5B, $Sb_2Te_3/Fe_{1+y}Te$ samples with 10-QL- and 24-QL-thick Sb_2Te_3 have very similar AFM transition temperatures, which indicates that the spin fluctuation in these 2 samples is nearly the same; however, the latter has a much higher onset SC temperature than the former. A similar circumstance also exists for the other 2 $Sb_2Te_3/Fe_{1+y}Te$ heterostructures with 15 QL and 27 QL of Sb_2Te_3 . Thus, the Sb_2Te_3 thickness dependence of the onset SC temperature revealed in Fig. 5A is a true property of these heterostructures. In SI Appendix, we describe an approach to fabricate 2 heterostructures with Sb_2Te_3 layers of different thicknesses grown on a single $Fe_{1+y}Te$ layer with a high spin fluctuation. The results of their SC characteristics (SI Appendix, Fig. S3) indeed provide further confirmation of this Sb_2Te_3 thickness dependence.

Spin-Orbit Coupling Alone Cannot Induce the Observed SC. In the following, we will briefly address the studies of another factor that was thought to be the underlying cause of the 2D SC in the

$Ti/Fe_{1+y}Te$ heterostructure system. It is related to an obvious and straightforward theoretical hypothesis that the strong spin-orbit coupling (SOC) of TI materials could itself induce the 2D SC. One can test this hypothesis experimentally by studying the transport properties of a $Fe_{1+y}Te$ heterostructure with a top layer possessing strong SOC but it is not a TI. We chose elemental Sb and Bi thin films as the replacement for the TI layers for the following reasons. The difference between A_2B_3 -type TI compounds and Sb is that the former compounds have a bulk energy gap with TSSs confined at the surfaces, while the latter is a semimetal (36, 37) and thus its TSSs penetrate to the entire interior of the film (38). On the other hand, due to an even stronger SOC in Bi (which is stronger than that of both Bi_2Te_3 and Sb_2Te_3), its surface bands inverse twice along $\bar{\Gamma}\bar{M}$ and return to the valence band, making Bi a semimetal with topologically trivial surface states (39) [except that ultrathin Bi films are a 2D TI (40)]. Thus, it is expected that both $Sb/Fe_{1+y}Te$ and $Bi/Fe_{1+y}Te$ heterostructures can be used to test if SOC alone could induce interfacial SC in $Fe_{1+y}Te$. SI Appendix, Fig. S4 displays the temperature-dependent resistance curves of 3 heterostructures, $Sb(10 \text{ nm})/Fe_{1+y}Te$, $Bi(13 \text{ nm})/Fe_{1+y}Te$, and $Sb_2Te_3(18 \text{ QL})/Fe_{1+y}Te$. As shown in this figure, the AFM transition temperatures for these 3 heterostructures are 54.8, 55.8, and 61.3 K, respectively, indicating that the AFM order of the $Sb_2Te_3/Fe_{1+y}Te$ heterostructure is stronger (equivalent to weaker spin fluctuation) than that of both the $Sb/Fe_{1+y}Te$ and $Bi/Fe_{1+y}Te$ heterostructures. The $Sb_2Te_3/Fe_{1+y}Te$ heterostructure is superconducting as expected; however, neither the

Fig. 5. The influences of Sb_2Te_3 thickness on the SC of $Sb_2Te_3/Fe_{1+y}Te$ heterostructures. (A) Normalized resistance as a function of temperature for $Sb_2Te_3/Fe_{1+y}Te$ heterostructures with varying thicknesses of the Sb_2Te_3 thin films of 10, 15, 24, and 27 QL. (Inset) The resistance at low temperature for the $Sb_2Te_3/Fe_{1+y}Te$ sample with 10 QL Sb_2Te_3 layer for clarifying the signature of its SC. (B) Normalized resistance for these 4 heterostructures in a zoomed-in region around the AFM transition. The arrows mark the AFM transition temperatures. The data plotted in A and B with colors in red, yellow, green, and blue are for $Sb_2Te_3/Fe_{1+y}Te$ heterostructures with the thickness of the top Sb_2Te_3 layer of 10, 15, 24, and 27 QL, respectively.

Sb/Fe_{1+y}Te nor the Bi/Fe_{1+y}Te heterostructure displays SC and they only show an up-turn in resistance at low temperature instead (*SI Appendix, Fig. S4*). These results indicate that SOC alone is not able to induce the interfacial SC observed in TI/Fe_{1+y}Te heterostructures.

Discussions

We believe that the observed Sb₂Te₃ thickness dependence on the SC quality of the heterostructure system described earlier can be linked to the evolution of the TSSs at the interface in the following ways. The degree of integrity (totality) of the TSSs at the interface increases with the Sb₂Te₃ thickness, which likely corresponds to higher SC quality of the heterostructure system. For thin Sb₂Te₃ layers, the 2 factors that can affect the degree of integrity of the interfacial TSSs are direct coupling and indirect coupling between the top and bottom TSSs. In direct coupling, the wave functions of the top and bottom TSSs are overlapped such that a small gap opens at the Dirac point, which reduces the integrity of the bottom TSSs. For Sb₂Te₃, the critical thickness for observing gapless TSSs is 4 QL, as demonstrated by scanning tunneling microscopy (41, 42) and ARPES (43). Since the observed Sb₂Te₃ thickness dependence in this study covers a thickness range from 10 to 27 QL, direct coupling should not play a key role in this dependence. Jiang et al. (44) previously reported that molecular beam epitaxy (MBE)-grown Sb₂Te₃ usually displays a p-type conductivity due to various intrinsic defects. We have also confirmed the p-type nature of our Sb₂Te₃ layers by conducting Hall measurements (*SI Appendix, Fig. S2*). This bulk conductivity was known to mediate a so-called indirect coupling between the top and bottom TSSs (45), which likely occurs even when the Sb₂Te₃ thickness is larger than the critical thickness of direct coupling. We believe this indirect coupling can also reduce the integrity of the TSSs at the interface of the heterostructure and the coupling is weakened as the TI thickness increases (46). Thus, indirect coupling is considered to be mainly responsible for the observed Sb₂Te₃ thickness dependence on the 2D SC of the heterostructure system. It is worth pointing out that such a TI-layer thickness dependence beyond the critical thickness for observing gapless TSSs of the Bi₂Te₃/Fe_{1+y}Te heterostructures is less obvious, as shown in our previous work (8). This can be explained by the big contrast between the estimated bulk carrier concentration of a Sb₂Te₃ (25 QL) and that of a Bi₂Te₃ (50 QL) thin film; the former is $1.63 \times 10^{22} \text{ cm}^{-3}$ (*SI Appendix, Fig. S2*), while the latter is about 1.3 to $1.6 \times 10^{19} \text{ cm}^{-3}$, as reported by us earlier (47). This big contrast implies that our Sb₂Te₃ thin film has a much higher bulk conductivity than Bi₂Te₃, leading to a stronger indirect coupling between the top and bottom TSSs for Sb₂Te₃ and, in consequence, a wider-ranging TI thickness dependence of the 2D SC as described above.

The fact that 2D SC of a Fe_{1+y}Te layer occurs only when it is covered with either Bi₂Te₃ or Sb₂Te₃ implies that a property of TI is likely involved in the induction of this SC. Since the TI layer in these heterostructures provides a source of itinerant carriers confined at the interface through the TSSs, it is possible that these itinerant carriers exhibit an unconventional pairing at the interface, leading to the observed interfacial SC. Recently, Yasuda et al. (48) observed a large nonreciprocal charge transport associated with the superconducting transition of Bi₂Te₃/FeTe, which is revealed to originate from the current-induced modulation of supercurrent density due to spin-momentum locking, indeed suggesting a close connection between SC and TSSs.

Below we briefly discuss the possible pairing mechanism of the 2D SC at the Sb₂Te₃/Fe_{1+y}Te interface. We believe the 2 key ingredients at the Sb₂Te₃/Fe_{1+y}Te interface are 1) 2D itinerant electrons induced by the TSSs of Sb₂Te₃ and 2) local spin moments with AFM ordering in Fe_{1+y}Te. The hybridization of the 2 ingredients, which are reminiscent of the Anderson lattice, was recently proposed to explain the strong superconducting pairing in

strongly correlated materials where inhomogeneous magnetism could appear as in electron-doped FeSe (49) and in spin-stripped nickelate (50); therefore, the possibility that spin order and SC could be intertwined cannot be ruled out. In fact, new spatial resolved methods show that the inhomogeneous charge-density-wave short-range order coexists with SC in cuprates (51). At the Sb₂Te₃/Fe_{1+y}Te interface, the fluctuation of the local moments of the AFM ordering in Fe_{1+y}Te is enhanced due to the hybridization with the itinerant electrons in TSSs. On the other hand, the enhanced fluctuating spin moments provide a driving force for the superconducting pairing of the itinerant electrons at the interface. The fact that SC transition temperature increases upon the increase of the fluctuation of the AFM order, as observed in this study, also implies that superconducting pairing benefits from the fluctuation of AFM order. Consequently, we propose that the superconducting pairing mediated by spin fluctuation enhanced by the hybridization of local spin moments in Fe_{1+y}Te and 2D itinerant electrons in the TSSs of Sb₂Te₃ is a possible mechanism of the SC observed in the Sb₂Te₃/Fe_{1+y}Te heterostructures. More quantitative experimental analysis and theoretical calculation are needed to confirm such a mechanism in future studies. We believe that the Sb₂Te₃/Fe_{1+y}Te heterostructures will provide a new platform not only for investigating the novel physics emerging from the interplay among SC, magnetism, and topological helical surface states but also for trapping and controlling Majorana fermions.

Methods

Sample Preparation. All of the samples studied in this work were fabricated by a VG-V80H MBE system equipped with in situ RHEED. Sample synthesis was conducted using high-purity Sb₂Te₃ and ZnSe compound sources together with Fe and Te elemental sources. A ZnSe buffer layer (80 nm) was firstly deposited on the GaAs (001) n+ substrates to form a flat surface, followed by a deposition of Fe_{1+y}Te with a thickness of 60 nm and a Sb₂Te₃ layer. For the studies of the interstitial Fe concentration dependence, a set of Sb₂Te₃ (24 QL)/Fe_{1+y}Te heterostructures and a set of pure Fe_{1+y}Te thin films with various amount of interstitial Fe in the Fe_{1+y}Te layer were fabricated by changing the temperature of Te effusion cell while keeping the temperatures of the Fe cell and substrate constant. Another set of Sb₂Te₃/Fe_{1+y}Te heterostructure samples were fabricated containing Sb₂Te₃ thin films with average thicknesses of 10, 15, 24, and 27 QL to study the Sb₂Te₃ thickness dependence of the 2D SC. All of the samples were capped with a Te protective layer (~8 nm) to avoid oxidation.

Characterizations. The thickness of each layer was determined by transmission electron microscope (TEM; JEOL 2010F). High-resolution spherical-aberration-corrected STEM images were carried out in a JEM-ARM200F TEM equipped with a probe corrector and a HAADF detector for finer crystal structure analysis. Sb₂Te₃/Fe_{1+y}Te heterostructures were cut into long strips (with dimension ~2 mm × 6 mm) and conventional 4-point electric contacts were made on the surface of each strip using silver paint as the contact material for performing ex situ electrical and magnetotransport measurements, which were carried out in a physical property measurement system equipped with a 14-T superconducting magnet. To determine the interstitial Fe concentration in the Fe_{1+y}Te layer, ex situ XPS measurements were performed by Kratos Axis Ultra DLD XPS equipped with a monochromatic Al K α X-ray source (photon energy 1,486.7 eV, 150 W), which was operated in hybrid lens mode with an energy step of 100.0 meV, a pass energy of 40 eV, and a large measuring area of 1 × 2 mm². Before acquiring a spectrum, Ar ion sputtering was performed to remove the upper layer(s) to reveal a pure surface of the Fe_{1+y}Te layer. The binding energy was calibrated with the C 1-s peak at 284.8 eV. The actual core-level peak areas were determined after applying Shirley background subtraction. High-resolution X-ray diffraction measurements were conducted using a PANalytical multipurpose diffractometer with an X'celerator detector (PANalytical X'Pert Pro) for crystalline characterizations.

Data Availability. All data included in this study are available via the 4TU.Centre for Research Data <https://researchdata.4tu.nl/en/> under the dataset identifier 18d09214-e75c-4846-bc53-73abe036f452.

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1. L. Fu, C. L. Kane, E. J. Mele, Topological insulators in three dimensions. *Phys. Rev. Lett.* **98**, 106803 (2007).
2. J. E. Moore, L. Balents, Topological invariants of time-reversal-invariant band structures. *Phys. Rev. B* **75**, 121306(R) (2007).
3. R. Roy, Topological phases and the quantum spin Hall effect in three dimensions. *Phys. Rev. B Condens. Matter Mater. Phys.* **79**, 195322 (2009).
4. H. J. Zhang *et al.*, Topological insulators in Bi_2Se_3 , Bi_2Te_3 and Sb_2Te_3 with a single Dirac cone on the surface. *Nat. Phys.* **5**, 438–442 (2009).
5. L. Fu, C. L. Kane, Superconducting proximity effect and majorana fermions at the surface of a topological insulator. *Phys. Rev. Lett.* **100**, 096407 (2008).
6. P. Zhang *et al.*, Observation of topological superconductivity on the surface of an iron-based superconductor. *Science* **360**, 182–186 (2018).
7. G. Wang *et al.*, Formation mechanism of superconducting $\text{Fe}_{1-x}\text{Te}/\text{Bi}_2\text{Te}_3$ bilayer synthesized via interfacial chemical reactions. *Cryst. Growth Des.* **14**, 3370–3374 (2014).
8. Q. L. He *et al.*, Two-dimensional superconductivity at the interface of a $\text{Bi}_2\text{Te}_3/\text{FeTe}$ heterostructure. *Nat. Commun.* **5**, 4247 (2014).
9. M. Q. He *et al.*, Pseudogap and proximity effect in the $\text{Bi}_2\text{Te}_3/\text{Fe}_{1-y}\text{Te}$ interfacial superconductor. *Sci. Rep.* **6**, 32508 (2016).
10. M. N. Kunchur *et al.*, Current-induced depairing in the $\text{Bi}_2\text{Te}_3/\text{FeTe}$ interfacial superconductor. *Phys. Rev. B Condens. Matter Mater. Phys.* **92**, 094502 (2015).
11. Q. L. He *et al.*, Anisotropic magnetic responses of a 2D-superconducting $\text{Bi}_2\text{Te}_3/\text{FeTe}$ heterostructure. *J. Phys. Condens. Matter* **27**, 345701 (2015).
12. C. L. Dean *et al.*, Current driven vortex-antivortex pair breaking and vortex explosion in the $\text{Bi}_2\text{Te}_3/\text{FeTe}$ interfacial superconductor. *Physica C* **527**, 46–49 (2016).
13. H. C. Liu *et al.*, Robust two-dimensional superconductivity and vortex system in $\text{Bi}_2\text{Te}_3/\text{FeTe}$ heterostructures. *Sci. Rep.* **6**, 26168 (2016).
14. K. Owada *et al.*, Electronic structure of $\text{Bi}_2\text{Te}_3/\text{FeTe}$ heterostructure: Implications for unconventional superconductivity. *Phys. Rev. B* **100**, 064518 (2019).
15. H. Qin *et al.*, Interfacial superconductivity induced by single-quintuple-layer Bi_2Te_3 on top of FeTe forming van-der-Waals heterostructure. arXiv:1903.06485 (15 March 2019).
16. E. E. Rodriguez *et al.*, Magnetic and structural properties near the Lifshitz point in Fe_{1+x}Te . *Phys. Rev. B Condens. Matter Mater. Phys.* **88**, 165110 (2013).
17. A. Bianconi, Shape resonances in superstripes. *Nat. Phys.* **9**, 536–537 (2013).
18. C. Koz *et al.*, Low-temperature phase diagram of Fe_{1+y}Te studied using x-ray diffraction. *Phys. Rev. B Condens. Matter Mater. Phys.* **88**, 094509 (2013).
19. T. J. Liu *et al.*, From $(\pi, 0)$ magnetic order to superconductivity with (π, π) magnetic resonance in $\text{Fe}_{(1.02)}\text{Te}_{(1-x)}\text{Se}_x$. *Nat. Mater.* **9**, 716–720 (2010).
20. C. de la Cruz *et al.*, Magnetic order close to superconductivity in the iron-based layered $\text{LaO}_{1-x}\text{F}_x\text{FeAs}$ systems. *Nature* **453**, 899–902 (2008).
21. S. Sanna *et al.*, Nanoscopic coexistence of magnetic and superconducting states within the FeAs layers of $\text{CeFeAsO}_{1-x}\text{F}_x$. *Phys. Rev. B* **82**, 060508(R) (2010).
22. R. S. Gonnelli *et al.*, Coexistence of two order parameters and a pseudogaplike feature in the iron-based superconductor $\text{LaFeAsO}_{1-x}\text{F}_x$. *Phys. Rev. B Condens. Matter Mater. Phys.* **79**, 184526 (2009).
23. J. E. Brom *et al.*, Structural and electrical properties of epitaxial Bi_2Se_3 thin films grown by hybrid physical-chemical vapor deposition. *Appl. Phys. Lett.* **100**, 162110 (2012).
24. J. Kampmeier *et al.*, Suppressing twin domains in molecular beam epitaxy grown Bi_2Te_3 topological insulator thin films. *Cryst. Growth Des.* **15**, 390–394 (2015).
25. F. Bonell *et al.*, Growth of twin-free and low-doped topological insulators on $\text{BaF}_2(111)$. *Cryst. Growth Des.* **17**, 4655–4660 (2017).
26. D. Kriegner *et al.*, Twin domain imaging in topological insulator Bi_2Te_3 and Bi_2Se_3 epitaxial thin films by scanning X-ray nanobeam microscopy and electron backscatter diffraction. *J. Appl. Crystallogr.* **50**, 369–377 (2017).
27. N. V. Tarakina *et al.*, Suppressing twin formation in Bi_2Se_3 thin films. *Adv. Mater. Interfaces* **1**, 1400134 (2014).
28. K. C. Kim *et al.*, Free-electron creation at the 60° twin boundary in Bi_2Te_3 . *Nat. Commun.* **7**, 12449 (2016).
29. J. Liang *et al.*, Dataset from “Studies on the origin of the interfacial superconductivity of $\text{Sb}_2\text{Te}_3/\text{Fe}_{1+y}\text{Te}$ heterostructures.” 4TU.Centre for Research Data. <https://data.4tu.nl/repository/uuid:18d09214-e75c-4846-bc53-73abe036f452>. Deposited 6 November 2019.
30. M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill Inc., 1996), pp. 93–94.
31. V. L. Berezinskii, Destruction of long-range order in one-dimensional and two-dimensional systems possessing a continuous symmetry group, II. Quantum systems. *Sov. Phys. JETP* **34**, 610–616 (1972).
32. B. I. Halperin, D. R. Nelson, Resistive transition in superconducting films. *J. Low Temp. Phys.* **36**, 599–616 (1979).
33. J. M. Kosterlitz, D. J. Thouless, Ordering, metastability and phase transitions in two-dimensional systems. *J. Phys. C Solid State Phys.* **6**, 1181–1203 (1973).
34. D. J. Scalapino, A common thread: The pairing interaction for unconventional superconductors. *Rev. Mod. Phys.* **84**, 1383–1417 (2012).
35. W. Bao *et al.*, Tunable (deltapi, deltapi)-type antiferromagnetic order in $\alpha\text{-Fe}(\text{Te}, \text{Se})$ superconductors. *Phys. Rev. Lett.* **102**, 247001 (2009).
36. D. Hsieh *et al.*, Observation of unconventional quantum spin textures in topological insulators. *Science* **323**, 919–922 (2009).
37. T. Kadono *et al.*, Direct evidence of spin-polarized band structure of $\text{Sb}(111)$ surface. *Appl. Phys. Lett.* **93**, 252107 (2008).
38. G. Yao *et al.*, Evolution of topological surface states in antimony ultra-thin films. *Sci. Rep.* **3**, 2010 (2013).
39. Y. M. Koroteev *et al.*, Strong spin-orbit splitting on bi surfaces. *Phys. Rev. Lett.* **93**, 046403 (2004).
40. I. K. Drozdov *et al.*, One-dimensional topological edge states of bismuth bilayers. *Nat. Phys.* **10**, 664–669 (2014).
41. T. Zhang, J. Ha, N. Levy, Y. Kuk, J. Stroscio, Electric-field tuning of the surface band structure of topological insulator Sb_2Te_3 thin films. *Phys. Rev. Lett.* **111**, 056803 (2013).
42. Y. Jiang *et al.*, Landau quantization and the thickness limit of topological insulator thin films of Sb_2Te_3 . *Phys. Rev. Lett.* **108**, 016401 (2012).
43. G. A. Wang *et al.*, Atomically smooth ultrathin films of topological insulator Sb_2Te_3 . *Nano Res.* **3**, 874–880 (2010).
44. Y. Jiang *et al.*, Fermi-level tuning of epitaxial Sb_2Te_3 thin films on graphene by regulating intrinsic defects and substrate transfer doping. *Phys. Rev. Lett.* **108**, 066809 (2012).
45. M. Brahlek, N. Koirala, M. Salehi, N. Bansal, S. Oh, Emergence of decoupled surface transport channels in bulk insulating $\text{Bi}_2(\text{S}, \text{Se})_3$ thin films. *Phys. Rev. Lett.* **113**, 026801 (2014).
46. W. J. Wang, K. H. Gao, Z. Q. Li, Thickness-dependent transport channels in topological insulator Bi_2Se_3 thin films grown by magnetron sputtering. *Sci. Rep.* **6**, 25291 (2016).
47. H. T. He *et al.*, Impurity effect on weak antilocalization in the topological insulator Bi_2Te_3 . *Phys. Rev. Lett.* **106**, 166805 (2011).
48. K. Yasuda *et al.*, Nonreciprocal charge transport at topological insulator/superconductor interface. *Nat. Commun.* **10**, 2734 (2019).
49. D. H. Lee, Routes to high-temperature superconductivity: A lesson from $\text{FeSe}/\text{SrTiO}_3$. *Annu. Rev. Condens. Matter Phys.* **9**, 261–282 (2018).
50. G. Campi *et al.*, Direct visualization of spatial inhomogeneity of spin stripes order in $\text{La}_{1.72}\text{Sr}_{0.28}\text{NiO}_4$. *Condens. Matter* **4**, 77 (2019).
51. G. Campi *et al.*, Inhomogeneity of charge-density-wave order and quenched disorder in a high- T_c superconductor. *Nature* **525**, 359–362 (2015).