Interfacial superconductivity in a bi-collinear antiferromagnetically ordered FeTe monolayer on a topological insulator

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The discovery of high-temperature superconductivity in Fe-based compounds triggered numerous investigations on the interplay between superconductivity and magnetism, and on the enhancement of transition temperatures through interface effects. It is widely believed that the emergence of optimal superconductivity is intimately linked to the suppression of long-range antiferromagnetic (AFM) order, although the exact microscopic picture remains elusive because of the lack of atomically resolved data. Here we present spin-polarized scanning tunnelling spectroscopy of ultrathin FeTe$_{1-x}$Se$_x$ ($x = 0, 0.5$) films on bulk topological insulators. Surprisingly, we find an energy gap at the Fermi level, indicating superconducting correlations up to $T_c \sim 6$ K for one unit cell FeTe grown on Bi$_2$Te$_3$, in contrast to the non-superconducting bulk FeTe. The gap spatially coexists with bi-collinear AFM order. This finding opens perspectives for theoretical studies of competing orders in Fe-based superconductors and for experimental investigations of exotic phases in superconducting layers on topological insulators.
Topological insulators (TIs) and interfacial superconductors (SCs) are both topics of intense current interest in modern condensed matter physics. The combination of both materials is expected to reveal novel physics such as, for example, Majorana Fermions arising in heterostructures of TIs and s-wave SCs by including magnetic fields. Most experimental studies of such heterostructures have concentrated on TI films grown on superconducting bulk substrates. Alternatively, SC/TI heterostructures could be realized by growing superconducting films on top of pristine bulk TIs.

In the past years, studies of Fe-based SCs and their electronic and magnetic properties have been one of the major research fields in condensed matter physics. The recent finding that the superconducting transition temperature $T_c$ can be pushed above 100 K in an FeSe unit cell (UC) thin film grown epitaxially on SrTiO$_3$ spurred numerous investigations aiming at an exact understanding of how the superconductivity in transition metal mono- and dichalcogenides evolves from bulk to ultrathin films. Bulk FeSe crystals exhibit a $T_c$ of no higher than 8 K (ref. 10). The significant interfacial enhancement of $T_c$ in UC FeSe on SrTiO$_3$ as well as in similar systems such as FeSe on BaTiO$_3$ (ref. 11) and FeTe$_{0.5}$Se$_{0.5}$ on SrTiO$_3$ (ref. 12) leads to a revival of the idea that tailoring the electron pairing by interfacing a SC with another material can be used to achieve high-temperature SC.

Interestingly, SC can even emerge by interfacing two insulators or a TI (Bi$_2$Te$_3$) with a non-SC Fe-chalcogenide (FeTe)$_{14,15}$. It is therefore of great importance to understand how different substrates affect the electronic, spin-dependent and superconducting properties of single-layer Fe-based SCs. The latter studies raised the fundamental question of what happens to the bi-collinear antiferromagnetic (AFM) structure of FeTe (ref. 16) once an interfacial SC state is established in contact with Bi$_2$Te$_3$. In the work of He et al. (ref. 14), it was proposed that the topological surface state may dope the FeTe and suppress the long-range AFM order at the interface, thereby inducing the observed interfacial SC.

By a low-temperature spin-polarized scanning tunnelling spectroscopy (SP-STS) investigation of the correlation between the film structure, the local electronic, superconducting and spin-dependent properties, we show here that, in contrast to this assumption, the energy gap at the Fermi level in UC thin FeTe films epitaxially grown on Bi$_2$Te$_3$(111) can spatially coexist on the atomic scale with long-range bi-collinear AFM order.

## Results

### Structural properties of FeTe$_{1-x}$Se$_x$ thin films.

We have prepared ultrathin epitaxial FeTe films on Bi$_2$Te$_3$ single crystals by in situ deposition of Fe on the TI substrates under ultrahigh vacuum (UHV) conditions, followed by an in situ annealing treatment (see Methods section). Figure 1a depicts a constant-current scanning tunnelling microscopy (STM) topographic image of such a film with FeTe islands of different thicknesses, together with regions of the bare substrate. We distinguish between these different areas based on the measured step heights and differences in the observed atomic-scale surface structures. The line profile displayed in Fig. 1b taken across three different terraces separated by two steps reveals a step height $h$ ($\alpha$–b) of $h \sim 0.65 \pm 0.05$ nm, roughly equal to the c axis lattice constant (0.62 nm) of bulk FeTe, while the right step b–c is considerably smaller ($h \sim 0.35 \pm 0.05$ nm). The smaller step results from an FeTe UC being embedded in the topmost quintuple layer of the substrate, most probably as sketched in Fig. 1b. Figure 1e presents an atomically resolved STM topographic image of the substrate area (Bi$_2$Te$_3$), which reveals the hexagonal close-packed atomic lattice (see inset Fast Fourier transform (FFT) image) of top Te atoms with a spacing of $\sim 0.44$ nm. A topographic STM image of the top UC layer of FeTe, with an atomic contrast determined by the topmost chalcogenide (Te) layer (Fig. 1f), reveals that the a axis of the FeTe is aligned parallel to one of the closed packed directions of the substrate Te atoms. The period of the atomic lattice along the a axis direction is $\sim 0.38$ nm, which is consistent...
with the Te–Te atomic distance on the surface of bulk FeTe (ref. 17). The period along the b axis is slightly smaller (a/b ~1.04), which might be partly attributed to the transition into the orthorhombic-monoclinic phase, known for bulk FeTe18, but could be enhanced by uniaxial strain as a result of the heteroepitaxial growth. This holds for the embedded UC FeTe as well as for thicker layers (Supplementary Fig. 1). However, there is no overall periodic height modulation of the UC FeTe layers, and only in some areas of the thinnest islands irregular wrinkles are observed. This indicates a rather weak interaction of the FeTe layer with the substrate, in contrast to what has been observed for FeSe grown on Bi2Se3 (ref. 19). The Fourier transform of the topographic image (inset of Fig. 1f) clearly shows the peaks associated with the almost square Te atomic lattice, with the spots along the a direction (q0.5) slightly more intense than those along the b direction (q1). Despite the different lattice symmetries (sixfold for Bi2Te3 and fourfold for FeTe) with respect to the c axis directions and the relatively large lattice mismatch, the heteroepitaxial growth is of very high quality, resulting in an atomically sharp and defect-free interface. In contrast to the surface of bulk FeTe (ref. 16), the surfaces of our ultrathin FeTe films do not show any excess Fe atoms, indicating a stoichiometric FeTe layer.

Although the focus of the present study is on the single layer of FeTe on Bi2Te2.5(111), ultrathin FeTe1-xSe0.5 (x = 0.5) films grown on the ternary TI Bi2Te1.8Se1.2(111) serve as an important reference system20. In the corresponding bulk system, FeTe exhibits a bi-collinear AFM order, while superconductivity can be induced and substantially enhanced up to 15 K with partial substitution of Se for Te at x = 0.5 (ref. 12). It is therefore particularly interesting to investigate how the superconducting state develops from a magnetically ordered state with Se substitution.

Characterization of superconducting properties. To characterize the electronic structure of these films, STS measurements of the differential tunnelling conductance (dI/dU) as a function of the applied bias voltage were performed, reflecting the sample’s local density of states. Figure 2a shows single tunnelling spectra measured on the one UC thin FeTe0.5Se0.5 film and on the Bi2Te2.5Se1.2 substrate at a temperature of T = 1.1 K. The FeTe0.5Se0.5 film exhibits an overall U-shaped dI/dU spectrum with an almost vanishing conductance value at the Fermi level (EF) and an energy gap of Δ = 2.5 meV, defined by half the distance between the two sharp coherence peaks in Fig. 2a. This measured gap value is of the same order of magnitude as measured on the surface of the corresponding superconducting bulk material23. In order to determine the critical temperature TC of our FeTe0.5Se0.5 films, we performed temperature-dependent measurements of dI/dU from 1.1 to 11 K (see Fig. 2b). As expected, the observed energy gap in dI/dU becomes shallower with increasing temperature and eventually disappears at ~11 K. The energy gap values for different temperatures (shown in Fig. 2c) were extracted from fits of the background-corrected and symmetrized dI/dU spectra (for data processing see Supplementary Fig. 3) employing the Dynes density of states, N(E) = N0(EF) · \frac{1}{\sqrt{|E - EF|^2 - \Delta^2(T^2)}} , with the density of states in the normal state at the Fermi energy N0(EF) and the real part of \gamma, where the parameter \gamma accounts for the lifetime broadening of the quasiparticles24. The fitting of the temperature dependence of the gap in the framework of the Bardeen–Cooper–Schrieffer (BCS) theory25 yields an energy gap at 0 K of Δ0 = 2.0 meV and an extracted TC = 11 K, similar to the values that have been extracted from STS on the corresponding superconducting bulk materials23,26. The temperature dependence of the dI/dU spectra as well as the derived TC value provide strong evidence that the observed gap feature arises from superconductivity in the ultrathin FeTe0.5Se0.5 film grown on Bi2Te2.5Se1.2.

Now we turn to the investigation of the electronic and magnetic structure of ultrathin FeTe films grown on Bi2Te3. Figure 2d shows a series of spatially averaged dI/dU spectra measured at T = 1.1 K on the Bi2Te3 substrate as well as on ultrathin FeTe films of different thicknesses. For both embedded UC and top UC FeTe layers a reduced dI/dU at EF with diffuse peaks at energies of ± 1 to ± 2 meV are found with a gap width and occurrence of the peaks varying with position (Supplementary Fig. 4). A gap of this size and temperature dependence (shown below) has not been observed at the surface of bulk FeTe (refs 27,28) or for FeTe single layers grown on SrTiO3 (ref. 12). The Fermi-level gap gradually vanishes for increased thicknesses of FeTe layers on Bi2Te3. For example, two UC thin FeTe films show a gap with a significantly reduced depth at EF. This behaviour suggests that the FeTe/Bi2Te3 interface plays a significant role for the observed gap structure. In order to reveal the origin of the energy gap in one UC thin FeTe films grown on Bi2Te3, we thoroughly investigated the temperature and field dependence of the dI/dU spectra. Figure 2e shows a series of background-corrected and symmetrized temperature-dependent dI/dU spectra for top UC FeTe on Bi2Te3. The tunnelling spectrum at T = 1.1 K exhibits a pair of diffuse peaks at U ~ 2 meV together with a gap region of reduced local density of states at EF. We observe that the gap depth and peak height in dI/dU decrease with increasing temperature and finally vanish above T = 6 K, very reminiscent of a superconducting transition. In order to test this hypothesis, the temperature-dependent spectra were fitted to a Dynes density of states (Fig. 2f). Kondo correlations, which were shown to cause dip features at EF for heavy Fermion systems29 and might be present on the Fe lattice as well, are ruled out, as they would show a different temperature dependence of the gap depth (Supplementary Fig. 6). In addition, unlike under-doped cuprates, where a pseudogap phase appears above Tc (ref. 30), the gap in our tunnelling spectroscopy data always closes at Tc ~ 6 K, thereby ruling out the presence of a pseudogap state. The spectral features therefore strongly suggest superconducting correlations of the UC FeTe layer. In order to extract the corresponding Tc, we fit the temperature dependence of the gap in the framework of the BCS theory (Fig. 2f), resulting in Tc ~ 6.5 K. Together with the zero temperature gap value of Δ0 = 1.05 meV extrapolated from the BCS fit we deduce a ratio of 2\Delta_0/k_B Tc ~ 3.8 for one UC thin FeTe on Bi2Te3, which is comparable to the value of 4.2 we determine for the FeTe0.5Se0.5 thin film on Bi2Te2.5Se1.2, but
somewhat larger than typical values of 2.5–3.2 for bulk FeTe₁₋ₓSeₓ samples. We note that the embedded UC of FeTe also shows a similar temperature-dependent behaviour (Supplementary Fig. 7). Our results are consistent with the transport measurements of Fe₈ +₅Te films grown on Bi₂Te₃ (ref. 15) where evidence for superconductivity with a Tc of about 8.9 Å at T = 1.1 K was reported. We do not observe any significant effect on the energy gap in a magnetic field up to B = 2.8 T applied perpendicular to the sample plane (Supplementary Fig. 8). This suggests that the robustness of the pairing with respect to an external magnetic field is similarly strong as for other Fe-based superconducting thin films, and an identical effect has been observed in magneto-transport measurements of a Bi₄Te₃/FeTe heterostructure.

**Spatial variation of superconducting correlation.** Having established clear evidence for superconducting correlations in single UC FeTe layers grown on Bi₂Te₃ below 6 K, we now focus in more detail on the spatial behaviour of these correlations at the lateral interface of the FeTe layer and the Bi₂Te₃ substrate. In particular, we measured the spatial variation of tunnelling spectra across a step edge from the embedded UC FeTe layer to the Bi₂Te₃ substrate (Fig. 3a–b). The resulting tunnelling spectra across the interface along the line shown in Fig. 3a obtained at T = 1.1 K (Fig. 3c) again show the presence of the gap due to superconducting correlations on top of the FeTe layer, which vanishes on the Tl substrate. To quantify the decay length of the superconducting correlations in our heterostructure system, we have fitted the spatial dependence of the gap area with an exponential decay function (Fig. 3d), resulting in a decay length of about 8.9 Å at T = 1.1 K. This decay length is a measure for the coherence length of the Cooper pairs, which is obviously smaller as, for example, in bulk FeTe₀.₈Se₀.₂ (ref. 26).

**Magnetic properties using SP-STM.** Our results on epitaxially grown FeTe layers on Bi₂Te₃ confirm that the previously reported
two-dimensional superconductivity in FeTe/Bi₂Te₃ heterostructures\textsuperscript{14,15} is associated with the presence of a superconducting layer of FeTe located at the interface. We now return to the central question of whether the usual bi-collinear AFM order of bulk FeTe (ref. 16) is suppressed in the FeTe layer interfacing Bi₂Te₃, as proposed by He et al.\textsuperscript{14}, or whether it can coexist with the superconducting correlations we observed.

In order to check this assumption, we simultaneously characterized the local superconducting correlations and the atomic-scale spin structure of the ultrathin FeTe films grown on Bi₂Te₃ by SP-STS\textsuperscript{33}. By using a spin-polarized bulk Cr tip\textsuperscript{34}, we excluded the disturbing influence of a local magnetic stray field on the sample while being sensitive to its out-of-plane surface spin component. Figure 4a represents the spin-resolved constant-current image of a single UC thin FeTe layer on Bi₂Te₃, as obtained by He et al.\textsuperscript{14}, or whether it can coexist with the superconducting correlations we observed.

Figure 3 | Spatial variation of energy gap across a lateral interface. (a) STM topography (25 nm × 25 nm) showing a Bi₂Te₃ terrace and an embedded UC thin FeTe layer grown on top (\(U = 100\) mV, \(I_s = 50\) pA). (b) Atomically resolved STM topography of a similar area (\(U = 50\) mV, \(I_s = 50\) pA, differentiated with respect to horizontal axis, white scale bar, 3 nm wide). (c) Two- and three-dimensional representation of normalized and symmetrized \(dI/dU\) spectra taken across the step along the line indicated in a in the direction of the arrow. (d) Evolution of the gap (markers), determined from the spectra in c by measuring the area enclosed by the gap within a ±1 mV voltage window. The solid line shows a fit to an exponential decay resulting in a decay length of \(\xi = 8.9\) Å. (e) Topographic height taken along the same line used for the data in c,d.
intensity of the spin contrast is almost the same for all three temperatures shown here. Similar SP-STM images at other temperatures have been taken in order to quantify the temperature evolution of the spin contrast via the ratio of FFT intensities of the \( q_{\text{AFM}} \) spots to the Bragg spots at \( q_{\text{Te}} \). Figure 5g shows this evolution together with the evolution of the superconducting energy gap taken from Fig. 2f. The comparison clearly demonstrates that the bi-collinear AFM order is largely unaffected by the disappearance of the superconducting correlations. Therefore, our experimental results do not provide any evidence that one kind of order emerges at the expense of the other, nor does the data provide any indication for a microscopic phase separation into regions with superconducting and magnetic order. Our findings for single UC thin FeTe layers on Bi$_2$Te$_3$ therefore challenge the common belief that optimal superconducting pairing sets in when long-range AFM order is suppressed in the parent compound.

**Discussion**

In summary, we have explored the electronic and magnetic structure in a new class of systems, that is, heterostructures consisting of ultrathin Fe-chalcogenide layers of the type FeTe$_{1-x}$Se$_x$ (\( x = 0, 0.5 \)) on Bi-based TI substrates. We observe fully developed U-shaped superconducting gaps in FeTe$_{0.5}$Se$_{0.5}$ layers of one UC thickness with a transition temperature of \( T_c \approx 11 \) K, close to the one of the corresponding bulk system (\( T_c \approx 14.5 \) K). For FeTe/Bi$_2$Te$_3$ heterostructures, our atomic-scale spin-resolved tunnelling spectroscopy measurements provide evidence for the coexistence of superconducting correlations with \( T_c \approx 6.5 \) K and bi-collinear AFM order in the one UC FeTe films. Although the coexistence of static magnetic order and superconductivity was observed earlier by spatially averaging techniques in several systems such as Fe-pnictides, heavy fermion compounds, and Fe-chalcogenides, the exact microscopic picture of the
robust and the intensity of the magnetic contrast is almost the same at all three temperatures. (with the experimental spectra. The errors in the magnetic contrast indicate the mean deviation as estimated from two data sets.

Figure 5 | Temperature stability of spin structure for top UC FeTe. (a–c) SP-STM images \((U = 100 \text{ mV} \text{ and } I_s = 100 \text{ pA})\), white scale bars, 3 nm wide, colour scale from 0 to 31 pm apparent height) and (d–f) corresponding FFT data (image size 0.75 Å⁻¹) acquired at \(B = 0.5 \text{ T}\) using an out-of-plane sensitive Cr tip, taken at different temperatures as indicated in a–c. The position of the surface area is the same in all three SP-STM data sets. The spin structure is robust and the intensity of the magnetic contrast is almost the same at all three temperatures. (g) Temperature dependence of the energy gaps \(\Delta\) plotted together with the magnetic contrast quantified via the ratio of intensities of the FFT spots at \(q_{\text{AFM}}\) and \(q_d\) from the SP-STM images in d–f and similar images taken at other temperatures. The errors in \(\Delta\) indicate the maximum range of values used in the Dynes fits, which result in an acceptable agreement with the experimental spectra. The errors in the magnetic contrast indicate the mean deviation as estimated from two data sets.

cocurrence still remained unclear because of a lack of spatially resolved data⁴⁰. Here we can compare the wavelength of the AFM order of \(\lambda = 2a = 7.5 \text{ Å}\) with the size of the Cooper pairs, inferred from the coherence length, of \(\xi = 8.9 \text{ Å}\), giving \(\xi/\lambda \sim 1.2\). The electron distance in the pairs is rather small, which is typical for Fe-based SCs, and apparently just large enough such that the effective Zeeman field induced by the AFM order cancels out along the length of the Cooper pair. The relative sizes of \(\lambda\) and \(\xi\) therefore might be crucial for the coexistence of pairing and long-range AFM order in this material. Our surprising findings may stimulate further theoretical studies on the relationship between superconductivity and AFM order in Fe-based SCs.

We finally note that the leakage of the gap into the TI substrate across the FeTe–Bi₂Te₃ interface (Fig. 3c) indicates the presence of superconducting correlations in the TI material close to the interface. The atomic sharpness of this interface suggests that the topological surface state of the TI substrate stays intact as also shown recently by photoemission experiments for the case of a FeSe–Bi₂Se₃ heterostructure⁴¹. The FeTe–Bi₂Te₃ interface therefore provides an ideal platform to study the interesting physics of Dirac fermions interacting with Cooper pairs.

Methods

Samples. Bulk TI single crystals of Bi₂Te₃ and Bi₂Te₃₋ₓSeₓ were used in this study as substrates. Fe deposited on Bi₂Te₃ reacts with the TI, and it is necessary to deposit Fe at low temperature to avoid Fe crystallization on the TI. The FeSe–Bi₂Se₃ heterostructure was prepared by depositing 0.5–1 ML Fe at 4 K. The FeTe–Bi₂Te₃ interface can be obtained by depositing 0.5–1 ML Fe at 300 K on top of Bi₂Te₃ and Bi₂Te₃₋ₓSeₓ substrates, respectively, using molecular beam epitaxy, followed by annealing up to a maximum temperature of 315 °C for 15 min. Fe deposited on Bi₂Te₃ reacts with the substrate upon annealing, most likely via a substitutional process of Bi by Fe (ref. 43), leading to a high-quality defect-free FeTe film.

Experimental techniques. All STM/STS experiments were performed with an STM in UHV at temperatures between 1.1 and 14 K (ref. 24). A magnetic field \(B\) of up to 3 T can be applied perpendicular to the sample surface. Topography images were obtained in constant-current mode with stabilization current \(I_s = 100 \text{ pA}\) and bias voltage \(V = 0.5 \text{ mV}\) applied to the sample. STS data were obtained using a lock-in technique to record the differential tunnelling conductance \(dI/dV\) by adding an AC modulation voltage \(U_{\text{mod}}\) (given in r.m.s.) to the bias voltage, after stabilizing the tip at \(I_s\) and \(V\), switching off the feedback, and ramping the applied bias \(U\). We used cut PtIr or electrochemically etched W tips (both in situ flashed) for spin-averaged imaging and spectroscopy measurements. For spin-resolved measurements, we used bulk Cr tips, which were prepared by electrochemical etching followed by a high-voltage field emission treatment using W(110) or Ta(001) as a substrate.

Data availability. The authors declare that the main data supporting the findings of this study are available within the article and its Supplementary Information files. Extra data are available from the corresponding author upon request.

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Author contributions

S.M., J.W., T.H. and R.W. designed the experiment. S.M., J.W., T.H. and R.W. performed the experiments. S.M., J.W., T.H. and R.W. analyzed the data. S.M., J.W., T.H. and R.W. wrote the manuscript. E.M.I.H., M.B., B.B.I. and Ph.H. have grown and characterized the TI single crystals. All authors discussed the results and contributed to the manuscript.

Additional information

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