



Pressure-induced reinforcement of interfacial superconductivity in a $\text{Bi}_2\text{Te}_3/\text{Fe}_{1+y}\text{Te}$ heterostructure



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ABSTRACT

We investigate the hydrostatic pressure dependence of interfacial superconductivity occurring at the atomically sharp interface between two non-superconducting materials: the topological insulator (TI) Bi_2Te_3 and the parent compound Fe_{1+y}Te of the chalcogenide iron-based superconductors. Under pressure, a significant increase in the superconducting transition temperature T_c is observed. We interpret our data in the context of a pressure-induced enhanced coupling of the Fe_{1+y}Te interfacial layer with the Bi_2Te_3 surface state, which modifies the electronic properties of the interface layer in a way that superconductivity emerges and becomes further enhanced under pressure. This demonstrates the important role of the TI in the interfacial superconducting mechanism.

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1. Introduction

Topological insulators (TI) have been a hot research topic since predicted and discovered in 2007 [1,2]. The novel three-dimensional TIs, e.g. Bi_2Te_3 and Bi_2Se_3 , own strong topological surface states with helical Dirac fermions, protected by time-reversal symmetry. Meanwhile, interfacial superconductivity also attracts intense interest from both theorists and experimentalists in various materials [3–5]. By combining a TI with an s-wave superconductor, the proximity effect at the surface states of TI has been studied, suggesting the existence of a topological superconductor, which may hold Majorana fermionic states [6]. This idea has triggered a search for Majorana fermions [7,8], which are of high interest for future applications in quantum computation [9].

We have recently reported a novel $\text{Bi}_2\text{Te}_3/\text{Fe}_{1+y}\text{Te}$ heterostructure, which exhibits interfacial superconductivity at an atomically-flat van-der Waals-bonded boundary between the non-superconducting parent compound Fe_{1+y}Te of the ‘11’ iron-based superconductor family and the non-superconducting topological insulator Bi_2Te_3 [10]. In this heterostructure, the TI surface state

is combined with the electronic complexity of the iron-based chalcogenide materials, resulting in a complex interfacial superconducting state. The interface exhibits all characteristic fingerprints of two-dimensional (2D) superconductivity with a Berezinskii–Kosterlitz–Thouless (BKT) transition below the resistive onset critical temperature (T_c^{onset}). In addition, a square-root or linear temperature dependence of the parallel and perpendicular upper critical field lines, respectively, was observed, which follow the 2D Ginzburg–Landau theory for a superconducting thickness of 7 nm. While a pure Fe_{1+y}Te film does not show any sign of superconductivity, with a Bi_2Te_3 film coated on the Fe_{1+y}Te the thin interfacial layer of the heterostructure becomes superconducting starting from one quintuple layer (QL) thickness of the TI. The T_c^{onset} increases with further Bi_2Te_3 thickness until it saturates at about 12 K for thicknesses exceeding 5 QLs. Since 5 QLs has been reported to represent the lower thickness limit of Bi_2Te_3 in order to fully develop a topological surface state [11,12], the dependence of T_c^{onset} on the number of QLs suggests that the TI surface state likely plays a crucial role for the emergence of the interfacial superconductivity. In addition, it has been reported that at the interface between Bi_2Te_3 and Fe_{1+y}Te the magnetic ordering of the latter compound is altered so that the iron magnetic moments order out of plane and thus perpendicular to the interface [13]. However, the superconducting mechanism remains unknown. The complexity of the superconducting gap structure, as observed in our previous point contact spectra [14] with multiple superconducting gaps, a pronounced pseudogap up to 30 K above T_c^{onset} , and

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a robust zero bias conductance peak, suggest a complex and peculiar superconducting pairing mechanism. In addition, it is unclear whether the highest critical temperature observed for more than 5 QLs thickness of Bi_2Te_3 can be further increased, for example by application of hydrostatic pressure.

High pressure has been known to be a powerful technique to induce superconductivity [15] and to modify the lattice structure or the charge carrier concentration [16,17]. This allows experiments in which an unconventional (e.g. iron-based) superconductor is tuned by pressure through its phase diagram using a single stoichiometric sample without introducing any crystalline disorder. In this article, we report data of electrical resistivity under the influence of hydrostatic (He-gas) pressure. A clear enhancement of both T_c^{onset} and the temperature T_0 where zero resistivity is established is observed in the resistance as the pressure increases up to 5.9 kbar. In addition, a sharp resistivity peak that occurs just above T_c^{onset} observed at ambient pressure is suppressed. The data are interpreted as a pressure-induced enhancement of the electronic interaction between the FeTe layer at the interface with the topological surface state of Bi_2Te_3 .

2. Experimental details

The heterostructure studied in this work was synthesized in a VG-V80H MBE system. A featured thickness of 9 quintuple layers (9 nm) of Bi_2Te_3 was capped onto the Fe_{1+y}Te layer (140 nm). The latter was grown on top of a ZnSe buffer layer (50 nm) deposited on the GaAs (001) semi-insulating substrates. Further details on the sample growth, morphology and characterization can be found in He et al. [10].

The electrical resistance measurements were conducted in a helium cryogenic Janis Supravertemp bath cryostat using the standard four-probe method with an AC current of $0.1 \mu\text{A}$ at 77 Hz. The sample voltage was amplified by a Stanford Research SR554 transformer preamplifier and fed into an SR830 DSP lock-in amplifier. Each sample was cut into the form of a long strip and silver paint was used to fabricate electrodes on the top surface of Bi_2Te_3 . Since the silver paint is known to quickly diffuse into Bi_2Te_3 [18,19], our electrodes should contact the entire TI layer, as well as a part of the Fe_{1+y}Te layer below. Thus, we are effectively probing both layers simultaneously.

To generate the pressure up to 7 kbar a He-gas compressor (Harwood Engineering) was connected to a CuBe helium gas pressure cell (Unipress, Warsaw) via a long capillary. With the sample sealed inside, the CuBe pressure cell was immersed in the liquid helium bath cryostat. The pressure values displayed in this article were measured using a digital manganin gauge at ambient temperature. At pressures below the freezing point of helium a correction was made using known helium isochores to obtain the slightly lower values of the pressure at 4 K. In this experiment the pressure was only changed at ambient temperature. Further details of the high-pressure experiment can be found in Hillier et al. [20].

3. Results

Fig. 1. shows the temperature-dependent resistance of the $\text{Bi}_2\text{Te}_3/\text{Fe}_{1+y}\text{Te}$ heterostructure (9 QLs) on a logarithmic temperature scale. Data at ambient pressure have been reported previously in Ref. [10,14]. Upon cooling from 295 K, the resistance is dominated by the bulk Fe_{1+y}Te , which effectively shunts the much thinner interface layer. It displays the characteristic insulator-to-metal transition in form of a maximum located at 76 K, characteristic for Fe_{1+y}Te with a rather high excess iron content [21]. This transition is well-known to originate from the antiferromagnetic double-stripe spin density wave (SDW) ordering in bulk Fe_{1+y}Te [22], and the corresponding characteristic temperature is noted as

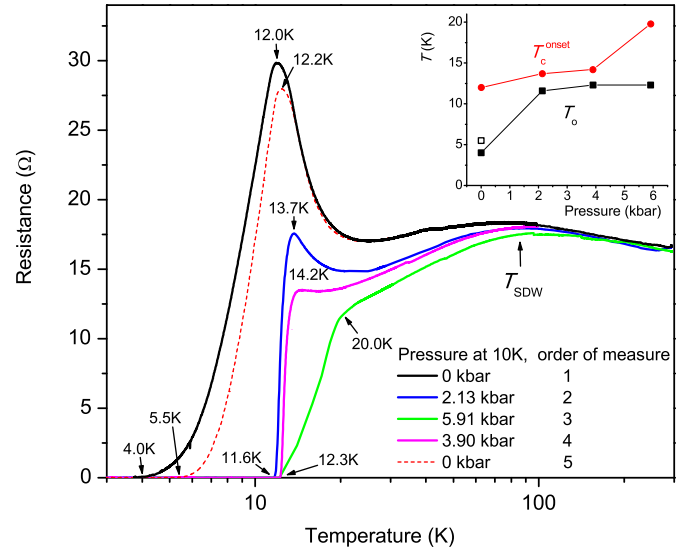


Fig. 1. The temperature dependent resistance measurement under pressures up to 5.91 kbar. The temperature T_0 where the resistance reaches zero is enhanced from 4.0 K to 12.3 K by applying pressure, while the peak located at T_c^{onset} is suppressed thus shifting the onset of the critical temperature up to 20 K at the highest pressure. T_{SDW} marks approximately the transition below which the antiferromagnetic spin density wave is formed. The inset shows the pressure evolution of the critical temperatures T_0 (where zero resistance is reached) and T_c^{onset} . The open square is T_0 after the pressure was released at the end of the experiment.

T_{SDW} . Note that Fe_{1+y}Te does not exist in a stoichiometric form (FeTe), but always contains excess iron in the form of interstitial ions. In our heterostructure the value of y is 0.15 ± 0.02 , as indicated by scanning transmission electron microscopy [10].

Upon further cooling the ambient pressure resistance goes through a minimum at 24 K, followed by a sudden increase with a sharp maximum at around 12 K, denoted as the onset T_c (T_c^{onset}), then a superconducting transition is clearly seen as the resistance drops gradually to zero at T_0 . This dramatic peak at T_c^{onset} is also typical for Fe_{1+y}Te with high excess iron [21], and likely originates from scattering on the interstitial iron magnetic moments in the bulk Fe_{1+y}Te layer. The superconducting transition has been demonstrated to be restricted to a 7-nm-thin planar region in the vicinity of the interface [10]. The resistance of the heterostructure is actually represented by three resistances in parallel: the thick bulk Fe_{1+y}Te layer, the interface layer and the Bi_2Te_3 layer. Above the superconducting transition onset the interface is normal conducting and the total resistance will be dominated by the much thicker bulk Fe_{1+y}Te layer. Below the superconducting onset the superconducting interfacial layer will then shunt the Fe_{1+y}Te and Bi_2Te_3 layers.

Upon application of pressure up to 5.9 kbar it is obvious that pressure decreases the overall Fe_{1+y}Te normal state resistance and suppresses the steep resistance increase just above T_c . In addition, the superconducting transition of the interface is clearly enhanced and both, T_c^{onset} and the zero resistance temperature T_0 are increased. This is illustrated in the inset of Fig. 1, where both T_c^{onset} and T_0 are plotted against the applied pressure. Note that pure Fe_{1+y}Te is not superconducting under hydrostatic pressure conditions [23], therefore the superconductivity is attributed at all pressures to the interface layer. It has been demonstrated previously that the finite transition width of the superconducting transition at ambient pressure is a consequence of the 2D nature of superconductivity, and well described by a BKT transition with additional finite size effects that can significantly alter the transition width [10]. In addition, the transition width increases drastically when the heterostructures are stored under protective nitrogen gas for

several months, which has been found to be intrinsic and not due to a deterioration in the sample quality [24]. This effect is likely a consequence of an ordering processes of the interstitial iron ions, and we will provide further evidence in this paper. The latter is the case for the investigated heterostructure, which enters a zero-resistance state only below $T_0 = 4\text{ K}$ where the superconductor becomes globally phase coherent. However, its normal state properties and the transition onset are identical to those previously reported [10,14]. The peak above T_c^{onset} is much lowered and almost flattened with a pressure of 3.9 kbar, then it disappears at 5.9 kbar together with a sudden displacement of T_c^{onset} up to 20 K. Meanwhile, T_0 increases to 11.6 K at a pressure of 2.13 kbar and reaches $T_0 = 12.3\text{ K}$ at 3.9 kbar.

4. Discussion

Above the superconducting transition temperature, the electrical transport of the heterostructure is largely dominated by the 140 nm thick layer of bulk Fe_{1+y}Te . The normal state resistivity at ambient pressure agrees well with literature data [10,14] for a similar large value of $y = 0.15 \pm 0.02$ as in our heterostructure, which shows a rather continuous insulator – metal transition in form of a resistivity maximum around 76 K at ambient pressure, instead of the sharper first-order resistivity jump for smaller y values. The upturn of the resistance below $\sim 20\text{ K}$, which leads to the peak just above T_c^{onset} , was also observed for such samples and attributed to the magnetic scattering of the charge carriers on the interstitial iron moments [21]. The paramagnetic high temperature insulating phase is almost unaffected by pressure, while the antiferromagnetically-ordered low-temperature metallic phase becomes more metallic. Due to the latter behavior, it is difficult to estimate the pressure dependence of T_{SDW} . It appears to be almost unaffected by pressure, although it has been reported that in bulk Fe_{1+y}Te the pressure decreases T_{SDW} [23].

For bulk Fe_{1+y}Te hydrostatic pressure is not known to induce superconductivity. In our heterostructure interfacial superconductivity with the maximum ambient pressure T_c occurs only in samples with a Bi_2Te_3 coating that exceeds the critical thickness reported to be required to completely develop the topological surface state [11,12]. The origin of the pressure-induced T_c enhancement is not to be expected in the FeTe layer alone, but it must include both the FeTe and Bi_2Te_3 interface layers. At the interface, the antiferromagnetic order of FeTe may be changed in a manner favorable for superconductivity. In addition, it has been reported that pressure has a strong influence on the topological surface states of Bi_2Te_3 [25], although it remains unclear how this can lead to an enhancement of the interfacial superconductivity. The main effect of hydrostatic pressure is certainly to push the different layers in the heterostructure closer together, thereby reducing the anisotropy and increasing the electronic coupling between the layers. This also applies to the coupling between the interfacial FeTe and Bi_2Te_3 layers, so that the influence of the surface state on the interface region can become more important. Although the mechanism of how this interaction with the surface state causes superconductivity remains unclear, a strengthening of the interaction may explain the T_c increase.

The pronounced peak above T_c^{onset} is quickly suppressed by application of pressure. The scattering of charge carriers on interstitial iron is therefore strongly suppressed and a more metallic trend emerges. We have previously shown that the interstitial iron couples magnetically to the iron moments in the Fe_{1+y}Te layers [26]. The disappearance of the peak suggests that the magnetic moments of the interstitial iron become more ordered, probably as a result of a stronger coupling to the antiferromagnetically ordered iron moments within the FeTe layers. Another possibility is that the pressure induces a spatial ordering, or a kind of clustering of

the interstitial iron. This is illustrated by the dramatic sharpening of the superconducting transition under pressure. The interstitial iron likely plays an important role in determining the normal conducting ground state of Fe_{1+y}Te . It provides strong pair breaking magnetic moments, and it also acts as a dopant [27]. Apart from this, a pressure-induced ordering can suppress the finite-size effect, which causes the broadening of the 2D Berezinski–Kosterlitz–Thouless transition [10]. The pressure-induced ordering or clustering of the interstitial iron could reduce the finite-size effect, resulting in the sharper transition. This is further supported by the fact that both the T_c^{onset} and T_0 remain higher than their initial values after releasing the pressure at the end of the experiment.

5. Conclusions

In summary, we measured the electrical resistivity on the novel interfacial superconductor $\text{Bi}_2\text{Te}_3/\text{Fe}_{1+y}\text{Te}$ under the influence of hydrostatic pressure. The temperature T_0 at which the zero resistance is established is increased from 4.0 K to 12.3 K with pressure rising from ambient conditions to 5.91 kbar. The observed pressure-induced increase in T_c is attributed to an enhancement of the electronic-coupling between the FeTe interface region and the topological Bi_2Te_3 surface state, although it remains unclear how the presence of the topological surface state causes the emergence of interfacial superconductivity. A local suppression of the antiferromagnetic SDW order that makes way to a superconducting ground state at the interface may represent a good candidate. In addition, we observed a dramatic sharpening of the resistive superconducting transition as well as a suppression of magnetic scattering above, ascribed to the pressure-induced ordering or clustering of interstitial iron in the Fe_{1+y}Te layer. Our experiments demonstrate the important role of the TI layer in the mechanism of interfacial superconductivity.

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References

- [1] L. Fu, C.L. Kane, Topological insulators with inversion symmetry, *Phys. Rev. B* 76 (2007) 045302.
- [2] M. König, S. Wiedmann, C. Brüne, A. Roth, H. Buhmann, L.W. Molenkamp, X.-L. Qi, S.-C. Zhang, Quantum spin hall insulator state in HgTe quantum wells, *Science* 318 (2007) 766–770.
- [3] A. Gozar, G. Logvenov, L. Fitting Kourkoutis, A.T. Bollinger, L.A. Giannuzzi, D.A. Muller, I. Bozovic, High-temperature interface superconductivity between metallic and insulating copper oxides, *Nature (London)* 455 (2008) 782–785.
- [4] H.Y. Hwang, Y. Iwasa, M. Kawasaki, B. Keimer, N. Nagaosa, Y. Tokura, Emergent phenomena at oxide interfaces, *Nat. Mater.* 11 (2012) 103–113.
- [5] S. Tan, Y. Zhang, M. Xia, Z. Ye, F. Chen, X. Xie, R. Peng, D. Xu, Q. Fan, H. Xu, J. Jiang, T. Zhang, X. Lai, T. Xiang, J. Hu, B. Xie, D. Feng, Interface-induced superconductivity and strain-dependent spin density waves in $\text{FeSe}/\text{SrTiO}_3$ thin films, *Nat. Mater.* 12 (2013) 634–640.
- [6] L. Fu, C.L. Kane, Superconducting proximity effect and Majorana fermions at the surface of a topological insulator, *Phys. Rev. Lett.* 100 (2008) 096407.
- [7] M. Veldhorst, M. Snelder, M. Hoek, T. Gang, V.K. Guduru, X.L. Wang, U. Zeitler, W.G. Van der Wiel, A.A. Golubov, H. Hilgenkamp, A. Brinkman, Josephson supercurrent through a topological insulator surface state, *Nat. Mater.* 11 (2012) 417–421.
- [8] F. Qu, F. Yang, J. Shen, Y. Ding, J. Chen, Z. Ji, G. Liu, J. Fan, X. Jing, C. Yang, L. Lu, Strong superconducting proximity effect in $\text{Pb-Bi}_2\text{Te}_3$ hybrid structures, *Sci. Rep.* 2 (2012) 339.
- [9] C. Nayak, S.H. Simon, A. Stern, M. Freedman, S.D. Sarma, Non-abelian anyons and topological quantum computation, *Rev. Mod. Phys.* 80 (2008) 1083–1159.

- [10] Q.L. He, H. Liu, M. He, Y.H. Lai, H. He, G. Wang, K.T. Law, R. Lortz, J. Wang, I.K. Sou, Two-dimensional superconductivity at the interface of a $\text{Bi}_2\text{Te}_3/\text{FeTe}$ heterostructure, *Nat. Commun.* 5 (2014) 4247.
- [11] Y.-Y. Li, G. Wang, X.-G. Zhu, M.-H. Liu, C. Ye, Y.-Y. Wang, K. He, L.-L. Wang, X.-C. Ma, H.-J. Zhang, X. Dai, Z. Fang, X.-C. Xie, Y. Liu, X.-L. Qi, J.-F. Jia, S.-C. Zhang, Q.-K. Xue, Intrinsic topological insulator Bi_2Te_3 thin films on Si and their thickness limit, *Adv. Mater.* 22 (2010) 4002–4007.
- [12] A.A. Taskin, S. Sasaki, K. Segawa, Y. Ando, Manifestation of topological protection in transport properties of epitaxial Bi_2Se_3 thin films, *Phys. Rev. Lett.* 109 (2012) 066803.
- [13] T. Hanke, U.R. Singh, L. Cornils, S. Manna, A. Kamlapure, M. Bremholm, E.M.J. Hedegaard, B. Brummerstedt Iversen, P. Hofmann, J. Hu, Z. Mao, J. Wiebe, R. Wiesendanger, Reorientation of the diagonal double-stripe spin structure at Fe_{1+y}Te bulk and thin-film surfaces, *Nat. Commun.* 8 (2017) 13939.
- [14] M. He, J. Shen, A.P. Petrovic, A.P. He, H.C. Liu, Y. Zheng, C.H. Wong, Q.H. Chen, J.N. Wang, K.T. Law, I.K. Sou, R. Lortz, Pseudogap and proximity effect in the $\text{Bi}_2\text{Te}_3/\text{Fe}_{1+y}\text{Te}$ interfacial superconductor, *Sci. Rep.* 6 (2016) 32508.
- [15] T. Park, E. Park, H. Lee, T. Klimczuk, E.D. Bauer, F. Ronning, J.D. Thompson, Pressure-induced superconductivity in CaFe_2As_2 , *J. Phys.* 20 (2008) 322204.
- [16] A. Eiling, J.S. Schilling, Evidence for Anderson lattice behavior in CeAg under pressure, *Phys. Rev. Lett.* 46 (1981) 364–367.
- [17] R. Lortz, A. Junod, D. Jaccard, Y. Wang, C. Meingast, T. Masui, S. Tajima, Evolution of the specific-heat anomaly of the high-temperature superconductor $\text{YBa}_2\text{Cu}_3\text{O}_7$ under the influence of doping through application of pressure up to 10 GPa, *J. Phys.* 17 (2005) 4135.
- [18] H.P. Dibbs, Thermal diffusion of silver in single-crystal bismuth telluride, *J. Appl. Phys.* 39 (1968) 2976–2977.
- [19] J. Keys, H. Dutton, Diffusion and solid solubility of silver in single crystal bismuth telluride, *J. Phys. Chem. Solids* 24 (1963) 563–571.
- [20] N.J. Hillier, N. Foroozani, D.A. Zocco, J.J. Hamlin, R.E. Baumbach, I.K. Lum, M.B. Maple, J.S. Schilling, Intrinsic dependence of T_c on hydrostatic (He gas) pressure for superconducting LaFePO , PrFePO , and NdFePO single crystals, *Phys. Rev. B* 86 (2012) 214517.
- [21] C. Koz, S. Rosler, A.A. Tsirlin, S. Wirth, U. Schwarz, Low-temperature phase diagram of Fe_{1+y}Te studied using x-ray diffraction, *Phys. Rev. B* 88 (2013) 094509.
- [22] W. Bao, Y. Qiu, Q. Huang, M.A. Green, P. Zajdel, M.R. Fitzsimmons, M. Zherenkov, S. Chang, M. Fang, B. Qian, E.K. Vehstedt, J. Yang, H.M. Pham, L. Spinu, Z.Q. Mao, Tunable $(\delta\pi, \delta\pi)$ -type antiferromagnetic order in $\alpha\text{-Fe}(\text{Te}, \text{Se})$ superconductors, *Phys. Rev. Lett.* 102 (2009) 247001.
- [23] Y. Mizuguchi, F. Tomioka, S. Tsuda, T. Yamaguchi, Y. Takano, FeTe as a candidate material for new iron-based superconductor, *Physica C* 469 (2009) 1027–1029.
- [24] H.-C. Liu, H. Li, Q.L. He, I.K. Sou, S.K. Goh, J. Wang, Robust two-dimensional superconductivity and vortex system in $\text{Bi}_2\text{Te}_3/\text{FeTe}$ heterostructures, *Scientific Reports* 6 (2016) 26168.
- [25] J. Liu, Y. Xu, J. Wu, B.-L. Gu, S.B. Zhang, W. Duana, Manipulating topological phase transition by strain, *Acta Cryst. C* 70 (2014) 118–122.
- [26] Q.L. He, M. He, J. Shen, Y.H. Lai, Y. Liu, H. Liu, H. He, G. Wang, J. Wang, R. Lortz, I.K. Sou, Anisotropic magnetic responses of a 2D-superconducting $\text{Bi}_2\text{Te}_3/\text{FeTe}$ heterostructure, *J. Phys.* 27 (2015) 345701.
- [27] L. Zhang, D.J. Singh, M.H. Du, Density functional study of excess Fe in Fe_{1+x}Te : magnetism and doping, *Phys. Rev. B* 79 (2009) 012506.