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To cite this article: Feifei Yuan *et al* 2018 *New J. Phys.* **20** 093012

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PAPER

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OPEN ACCESS

RECEIVED

23 May 2018

REVISED

26 July 2018

ACCEPTED FOR PUBLICATION

3 September 2018

PUBLISHED

12 September 2018

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Keywords: Fe-based superconductors, thin film, upper critical fields

Abstract

Revealing the universal behaviors of iron-based superconductors (FBS) is important to elucidate the microscopic theory of superconductivity. In this work, we investigate the effect of in-plane strain on the slope of the upper critical field H_{c2} at the superconducting transition temperature T_c (i.e. $-dH_{c2}/dT$) for FeSe_{0.7}Te_{0.3} thin films. The in-plane strain tunes T_c in a broad range, while the composition and disorder are almost unchanged. We show that $-dH_{c2}/dT$ scales linearly with T_c , indicating that FeSe_{0.7}Te_{0.3} follows the same universal behavior as observed for pnictide FBS. The observed behavior is consistent with a multiband superconductivity paired by interband interaction such as sign change s_{\pm} superconductivity.

1. Introduction

The discovery of superconductivity in the iron oxypnictide LaFeAs(O,F) has triggered a surge of research on Fe-based superconductors (FBS) [1–4]. Systematic studies revealed several universal scaling behaviors for FBS as for example: (1) the Bud'ko–Ni–Canfield (BNC) scaling of the specific heat jump ΔC at the superconducting transition temperature T_c with $\Delta C \propto T_c^3$ for the majority of FBS [5–11], (2) the linear dependence of the slope of the upper critical field $-dH_{c2}/dT$ at T_c versus T_c [12, 13], and (3) the relations between T_c and the structural parameters such as the As–Fe–As bond angle and the anion height [14, 15]. So far, there is no general agreement on the interpretation of these scaling behaviors. Kogan *et al* proposed that the BNC scaling $\Delta C \propto T_c^3$ and $-dH_{c2}/dT \propto T_c$ is related to an intrinsic pair-breaking in superconductors with strongly anisotropic order parameters, such as FBS [12, 16]. Alternatively, Zaanen *et al* discussed the idea that BNC scaling is expected for a quantum critical metal undergoing a pairing instability [17, 18]. Moreover, Bang *et al* pointed out that the observed scaling behaviors can be a generic property of the multiband superconducting state paired by a dominant interband interaction [19, 20].

In the case of FeSe_{1-x}Te_x, the universal scaling behavior was found for the anion height position [14] and for ΔC [11]. However, it has not been reported for H_{c2} so far. Recently, we demonstrated that biaxial in-plane strain allows to change T_c of the FeSe_{1-x}Te_x thin films with $x \approx 0.3$ in a broad temperature range avoiding phase separation [21]. This allows to study the behavior of H_{c2} at well-defined conditions. In this work by measuring the electrical resistance in magnetic field we show that also FeSe_{0.7}Te_{0.3} thin films follow the universal scaling behavior of $-dH_{c2}/dT \propto T_c$ in a broad range of T_c , as observed in LnOFeAs (Ln: lanthanoid elements) and AEF₂As₂ (AE: alkaline earth elements) compounds [12, 13].

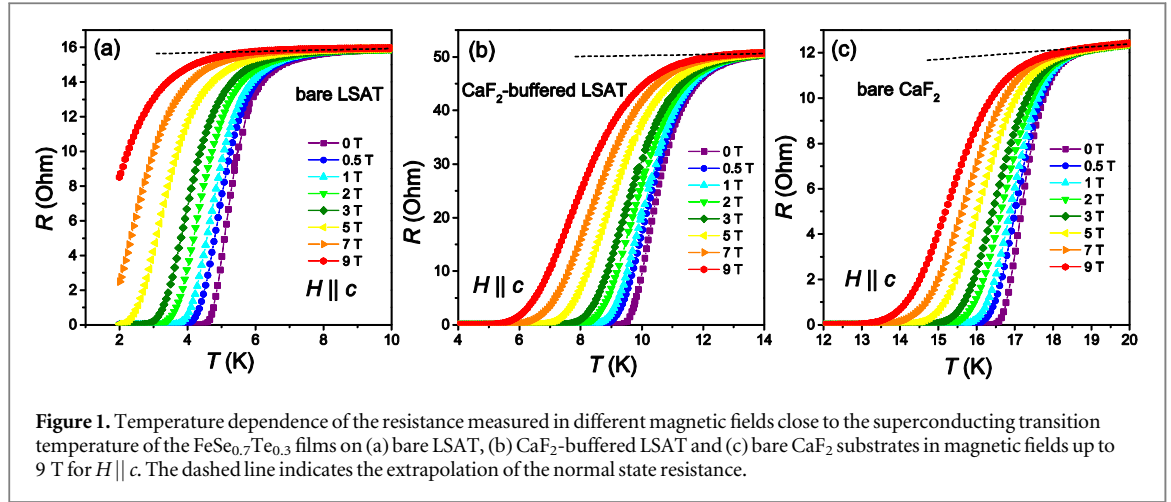


Figure 1. Temperature dependence of the resistance measured in different magnetic fields close to the superconducting transition temperature of the $\text{FeSe}_{0.7}\text{Te}_{0.3}$ films on (a) bare LSAT, (b) CaF_2 -buffered LSAT and (c) bare CaF_2 substrates in magnetic fields up to 9 T for $H \parallel c$. The dashed line indicates the extrapolation of the normal state resistance.

Table 1. Structural and superconducting properties of the films on different substrates presented in this paper.

| Substrate | a (Å) | 90% | | | 50% | | |
|------------------------|---------|-----------|-----------------|------------------------------|-----------|-----------------|------------------------------|
| | | T_c (K) | $H_{c2}(0)$ (T) | $-dH_{c2}/dT$ (T K $^{-1}$) | T_c (K) | $H_{c2}(0)$ (T) | $-dH_{c2}/dT$ (T K $^{-1}$) |
| LSAT | 3.787 | 6.35 | 17.11 | 2.69 | 5.27 | 11.99 | 2.27 |
| | 3.788 | 7.39 | 24.83 | 3.36 | 5.74 | 14.66 | 2.55 |
| | 3.777 | 12.23 | 76.79 | 6.28 | 10.96 | 43.47 | 3.96 |
| CaF_2 -buffer | 3.776 | 11.93 | 59.37 | 4.98 | 10.53 | 34.38 | 3.27 |
| | 3.775 | 13.28 | 88.74 | 6.68 | 11.94 | 49.52 | 4.15 |
| CaF_2 | 3.748 | 19.11 | 158.29 | 8.29 | 18.62 | 98.99 | 5.32 |
| | 3.753 | 19.31 | 139.54 | 7.23 | 18.42 | 81.47 | 4.42 |
| | 3.761 | 18.09 | 144.58 | 7.99 | 17.22 | 81.71 | 4.74 |

2. Experiment

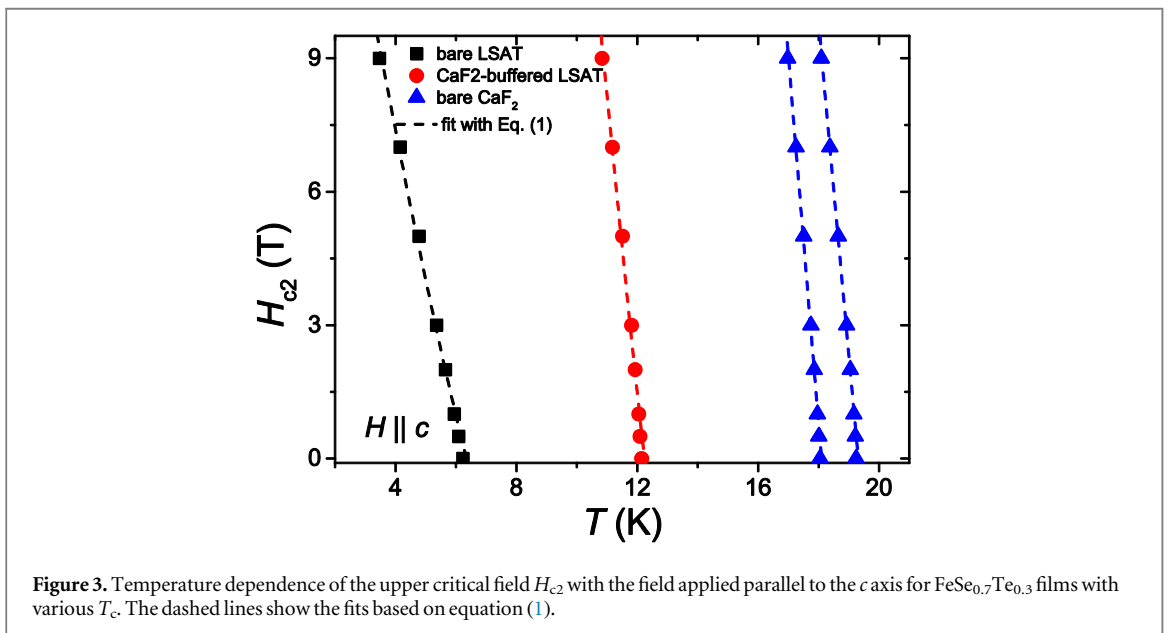
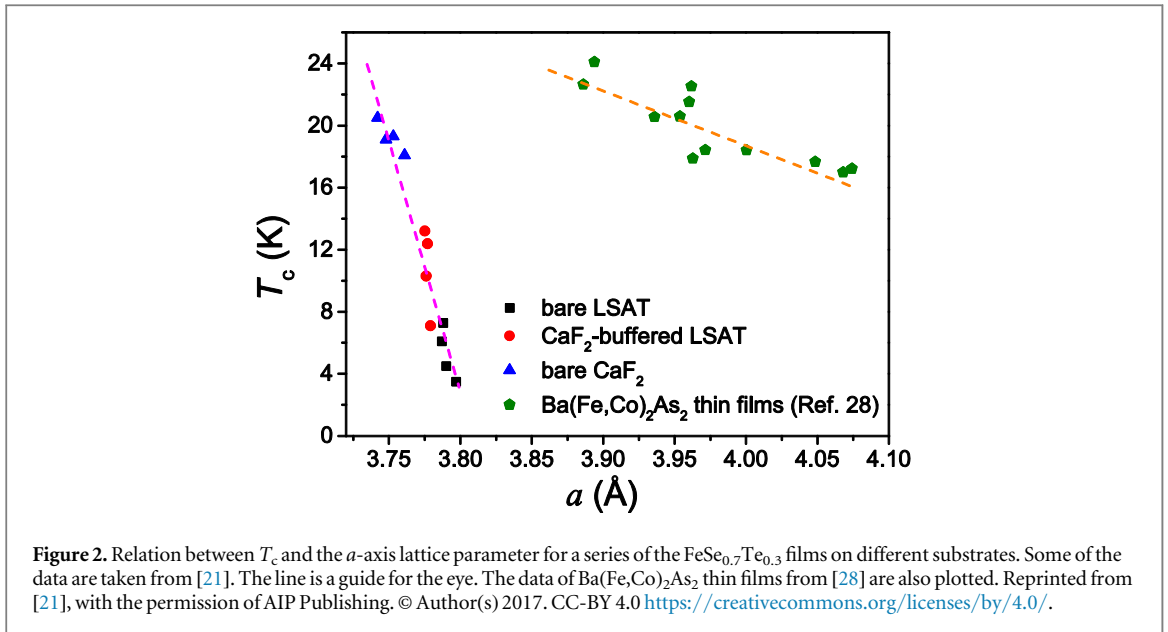
The thin films were prepared starting from a stoichiometric $\text{FeSe}_{0.5}\text{Te}_{0.5}$ target on various substrates, namely $(\text{La}_{0.18}\text{Sr}_{0.82})(\text{Al}_{0.59}\text{Ta}_{0.41})\text{O}_3$ (LSAT), CaF_2 -buffered LSAT, and bare CaF_2 (001)-oriented single crystalline substrates using pulsed laser deposition with a KrF excimer laser (wavelength: 248 nm, repetition rate: 7 Hz) under ultrahigh vacuum conditions with a background pressure of 10^{-9} mbar [21, 22].

The lattice parameter a was derived from reciprocal space maps measured in a PANalytical X'pert Pro system. Transmission electron microscopy (TEM) investigations of the films were performed in a FEI Tecnai-T20 TEM operated at 200 kV acceleration voltage. TEM lamellae were prepared by a focused ion beam technique (FIB) in a FEI Helios 600i using an acceleration voltage of 3 kV in the last FIB step. The composition of the samples was determined by energy-dispersive x-ray spectroscopy (EDX) with an Edax EDAMIII spectrometer in TEM. EDX line scans across the cross-section of the films confirmed the stoichiometry to be homogeneous over the film thickness, as shown in figure A1 in the appendix. It was found that the composition of the films is $\text{FeSe}_{0.7}\text{Te}_{0.3}$ within the error-bars of the analysis (few percent) for all studied substrates due to the preference of Fe to bond with Se because of the low formation energy [23]. Electrical transport properties were measured in a Physical Property Measurement System [(PPMS) Quantum Design] by a standard four-probe method, for which 4 pins are collinearly aligned along the edge of the film. More details on these structural properties are found in a recent publication of our group [21].

3. Results and discussion

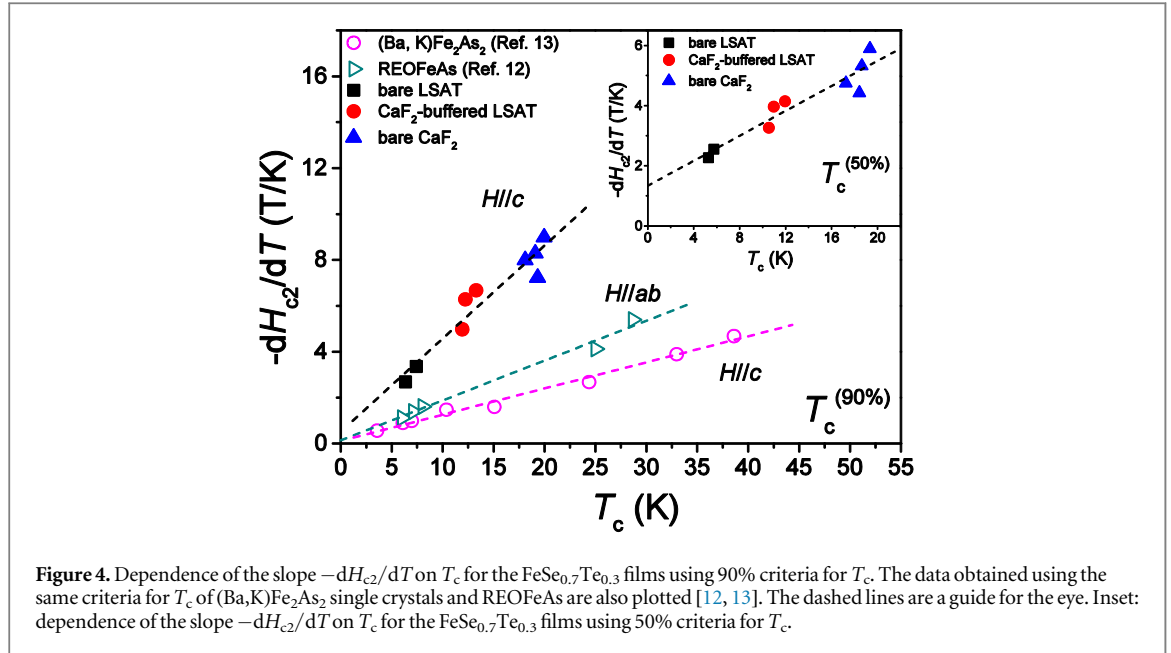
3.1. Effect of strain on T_c

The temperature dependence of the resistance is shown in figure 1 for $\text{FeSe}_{0.7}\text{Te}_{0.3}$ films grown on different substrates (i.e. bare LSAT, CaF_2 -buffered LSAT and bare CaF_2) measured in magnetic fields up to 9 T for $H \parallel c$. The dashed lines indicate the fit of the normal state just above the superconducting transition temperature T_c by $R(T) = R_0 + AT$, where R_0 is the resistance extrapolated to $T = 0$ K (i.e. residual resistance) and A is a constant. T_c is defined as 90% of the resistance in the normal state. As shown in figure 1(a), the lowest T_c of 6.2 K without magnetic field is measured for the films on bare LSAT substrate. T_c is nearly double by employing a 25 nm CaF_2



buffer layer (figure 1(b)). Furthermore, the $\text{FeSe}_{0.7}\text{Te}_{0.3}$ thin films on bare CaF_2 substrate have the highest $T_c \sim 18.1$ K, shown in figure 1(c) (the data for additional films can be found in figure A3 in the appendix). The applied magnetic field suppresses T_c resulting in a monotonous broadening of the transition attributed to different temperature dependencies of H_{c2} and irreversibility fields (H_{irr}). The temperature dependence of the normalized resistance for different films is shown in figure A2. The value of the residual resistivity ratio defined by $R(300 \text{ K})/R(20 \text{ K})$ is consistent with the results reported by other groups [24, 25], and is nearly substrate independent.

As mentioned above, the particularities of the crystal structure have a strong effect on T_c in FBS [26, 27]. Recently we found that T_c of the $\text{FeSe}_{0.7}\text{Te}_{0.3}$ films is very sensitive to in-plane lattice parameter a [21]. T_c as a function of the a -axis is shown in figure 2 for a number of $\text{FeSe}_{0.7}\text{Te}_{0.3}$ films grown on the mentioned substrates. For comparison, values of optimally doped $\text{Ba}(\text{Fe}_{0.92}\text{Co}_{0.08})_2\text{As}_2$ thin films on different substrates are also plotted [28]. The films have different a -axis lengths due to different in-plane compressive strain resulting mainly from the large thermal misfit between the substrates and the $\text{FeSe}_{1-x}\text{Te}_x$ layer [21, 22, 28–32]. It is apparent that the superconducting transition temperature T_c decreases linearly with increasing a -axis lattice parameter for the $\text{FeSe}_{0.7}\text{Te}_{0.3}$ films, which is consistent with reports of other groups [33–35]. A linear dependence of T_c on the crystallographic a -axis was also found for $\text{Ba}(\text{Fe},\text{Co})_2\text{As}_2$ thin films [28]. However, $\text{FeSe}_{0.7}\text{Te}_{0.3}$ has a much steeper slope. The high sensitivity to strain in the FeSe system can be attributed to the presence of shallow Fermi pockets (with small Fermi energy ϵ_F) [34]. The strain shifts slightly the position of the bands with respect to the



chemical potential resulting in a considerable change of the small ε_F value or even the appearance of a Lifshitz transition [34]. These changes of the electronic structure can affect T_c as was demonstrated before for the 122 system [30]. This allows us to vary T_c of the $\text{FeSe}_{0.7}\text{Te}_{0.3}$ films significantly solely by in-plane strain.

3.2. Upper critical fields near T_c

Figure 3 shows H_{c2} for the films on different substrates as a function of T_c for fields parallel to the c axis (the data for additional films can be found in figure A3(e) in the appendix) with 90% criteria for T_c . To describe the H_{c2} curves for the $\text{FeSe}_{0.7}\text{Te}_{0.3}$ compound, the spin paramagnetic and the orbital pair-breaking effects should be taken into account [36–38]. A commonly used approach is the WHH theory generalized for multiband superconductors [39]. However, for the reliable determination of the paramagnetic and multiband effects, this analysis requires H_{c2} values in a broad temperature range. Due to very high H_{c2} values of this compound, only the data near T_c are accessible in our experiments. Therefore, we used alternatively an analysis based on the Ginzburg–Landau theory, which provides a simple analytical expression for the temperature dependence of $H_{c2}(T)$ near T_c [40–42]. H_{c2} and its slope near T_c can be estimated by the following equation:

$$H_{c2} = H_{c2}(0) \left[\frac{1 - t^2}{1 + t^2} \right], \quad (1)$$

where $t = T/T_c$ is the reduced temperature and $H_{c2}(0)$ is the upper critical field values extrapolated to $T = 0$. We fitted our H_{c2} data using equation (1), as shown by the dashed line in figure 3. The slope $-dH_{c2}/dT$ at T_c is defined using equation (1) at $t = 1$ (figure 4). The analysis including paramagnetic effects results in quantitative changes of the slope (see figure A4 in the appendix) [43]. However, the functional dependence of the slope on T_c is qualitatively unchanged (figure 4 below and figure A4) in spite of relatively strong paramagnetic effects with the Maki parameter $\alpha_M = \sqrt{2}H_{c2}^*/H_p$ given in inset of figure A4 in the appendix, where H_{c2}^* is the orbital limited upper critical field and H_p is the paramagnetic critical field. We found also that $\alpha_M \propto T_c$ in accord with the scaling behavior of H_{c2} discussed in the next section. The large values of the α_M are consistent with previous studies of the $\text{FeSe}_{1-x}\text{Te}_x$ system [44].

3.3. The slope of H_{c2} at T_c interpreted by a single-band model

The derivative of the upper critical field with temperature $-dH_{c2}/dT$ versus T_c is shown in figure 4. As can be seen, the slope $-dH_{c2}/dT$ of $\text{FeSe}_{0.7}\text{Te}_{0.3}$ thin films depends almost linearly on T_c in accordance with the behavior $-dH_{c2}/dT \propto T_c$ found in some other pnictides [12, 13]. We note that the choice of criteria does not change the observed linear dependence qualitatively. The $-dH_{c2}/dT$ data using 50% of the resistance in the normal state as the criterion of T_c is shown in the inset of figure 4. In contrast to the 90% criterion, the slope does not reach zero by an extrapolation to $T_c = 0$, which indicates that $-dH_{c2}/dT$ is affected by additional T_c independent contributions (such as sample inhomogeneity) not related to H_{c2} . Therefore, for further analysis we focused on the data obtained with the 90% criterion.

A linear behavior of $-dH_{c2}/dT \propto T_c/\nu_F^2$ is expected for a single-band superconductor in the clean limit, if the Fermi velocity (ν_F) is the same for different samples [39]. For our samples, the linear relation between T_c and

lattice parameters (figure 2) indicates that the mechanism for a T_c suppression is related to the modification of the electronic properties, which should result in a variation of ν_F with strain. Therefore, for a clean limit we expect a deviation from the linear behavior $-dH_{c2}/dT \propto T_c$. In a dirty limit, the slope $-dH_{c2}/dT \propto 1/D$ of an s -wave superconductor is independent on T_c , where D is an effective diffusivity constant [45]. It is known that T_c of a single-band s -wave superconductor can be suppressed by magnetic impurities. However, smaller D values (stronger impurity scattering) result in a lower T_c and higher $-dH_{c2}/dT$ in contrast to the experimental observations. Therefore, for our films effectively a single-band s -wave superconductivity cannot reconcile the whole set of the experimental data.

3.4. The slope of H_{c2} at T_c interpreted by a two-band model

In the case of superconductivity driven by a single leading interband interaction such as s_{\pm} , the minimal model that describes the system close to T_c is a two-band model [46]. The most of available experimental data are consistent with this picture. So far we have found that only hole overdoped $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ system that breaks time reversal symmetry in the superconducting state cannot be described by a two-band model [47]. In this case, a three-band model is needed to describe the superconducting properties [48]. However, general scaling behaviors do not hold in this case (see introduction in [47]). Therefore, the observed universality in scaling behavior of H_{c2} relates our $\text{FeSe}_{0.7}\text{Te}_{0.3}$ films to the majority FBS with superconductivity driven by a single leading interband interaction.

It was shown that the pair-breaking parameters do not alter the slope for dirty superconductors with a sign changed order parameter such as s_{\pm} superconductors and for two symmetrical bands the slope is $-dH_{c2}/dT \propto T_c / \langle W^2 \nu_F^2 \rangle$, where Ω is the variation of the gap along the Fermi surface [12, 16]. The symmetrical case can be excluded due to the expected variation of the Fermi velocity with strain, which results in a deviation from the linear behavior. The symmetrical case is also inconsistent with the available angle-resolved photoemission spectroscopy (ARPES) data [49]. The effect of impurities on a realistic s_{\pm} superconductor (non-symmetrical bands and non-zero intraband coupling) is rather complex and results for strong enough impurities in a transition to the s_{++} superconducting state [50]. Therefore, an interpretation of the observed linear behavior based on a strong pair breaking effect is also doubtful for the 11 system.

In a clean two-band s -wave superconductor, the slope of H_{c2} is defined by a combination of the Fermi velocities and coupling constants for different bands:

$$-\left. \frac{dH_{c2}}{dT} \right|_{T_c} \propto \frac{T_c}{(a_1 v_1^2 + a_2 v_2^2)}, \quad (2)$$

where $a_1 = \frac{\sqrt{(\lambda_{11} - \lambda_{22})^2 + 4\lambda_{12}\lambda_{21}} + \lambda_{11} - \lambda_{22}}{2(\lambda_{11}\lambda_{22} - \lambda_{12}\lambda_{21})}$ and $a_2 = \frac{\sqrt{(\lambda_{11} - \lambda_{22})^2 + 4\lambda_{12}\lambda_{21}} - \lambda_{11} + \lambda_{22}}{2(\lambda_{11}\lambda_{22} - \lambda_{12}\lambda_{21})}$ are constants, which depend on the intraband λ_{11} , λ_{22} and interband λ_{12} , λ_{21} coupling constants [39]. $a_1 \sim a_2$ holds in the case of an extreme s_{\pm} superconductivity with the dominant interband coupling, which presumably is the case for $\text{FeSe}_{1-x}\text{Te}_x$ [51, 52], and $a_1 \gg a_2$ for an extreme s_{++} case with the dominant intraband coupling. The latter can be excluded based on the arguments for a single-band case since the leading band dominates the superconducting properties close to T_c . In the case of strong interband coupling, the universal behavior $-dH_{c2}/dT \propto T_c$ would indicate that the combination $a_1 v_1^2 + a_2 v_2^2$ is nearly strain independent. It is known that the value of the Fermi velocities considerably varies between different bands in the 11 system [49]. Therefore, according to equation (2), $-dH_{c2}/dT$ is dominated by the fastest Fermi velocity assuming sizeable interband coupling, which is expected in the case of the strongly anisotropic sign change superconducting gap [50, 51] or special s_{++} case [53]. The linear scaling indicates that the fastest Fermi velocity is weakly sensitive to strain assuming a weak variation of the coupling constants with strain. In this case, T_c is mainly defined by the band/bands with low Fermi velocities forming small Fermi surface pockets. This is consistent with empirical conclusions based on the ARPES measurements of various pnictides [54]. The universality of the observed scaling $-dH_{c2}/dT \propto T_c$ for different FBS imposes constrain on the possible pairing mechanism and indicates a key role of the interband interactions.

4. Summary

The superconducting transition temperature of $\text{FeSe}_{0.7}\text{Te}_{0.3}$ films can be significantly modified solely by in-plane biaxial strain. We observed that the slope of the upper critical field of the strained films is proportional to T_c as found for other classes of FBS materials. The behavior observed indicates a striking similarity in the nature of superconducting state between the $\text{FeSe}_{1-x}\text{Te}_x$ system and iron pnictides. This also suggests that the behavior $-dH_{c2}/dT \propto T_c$ may be a generic property of multiband superconductors paired by a dominant interband pairing potential.

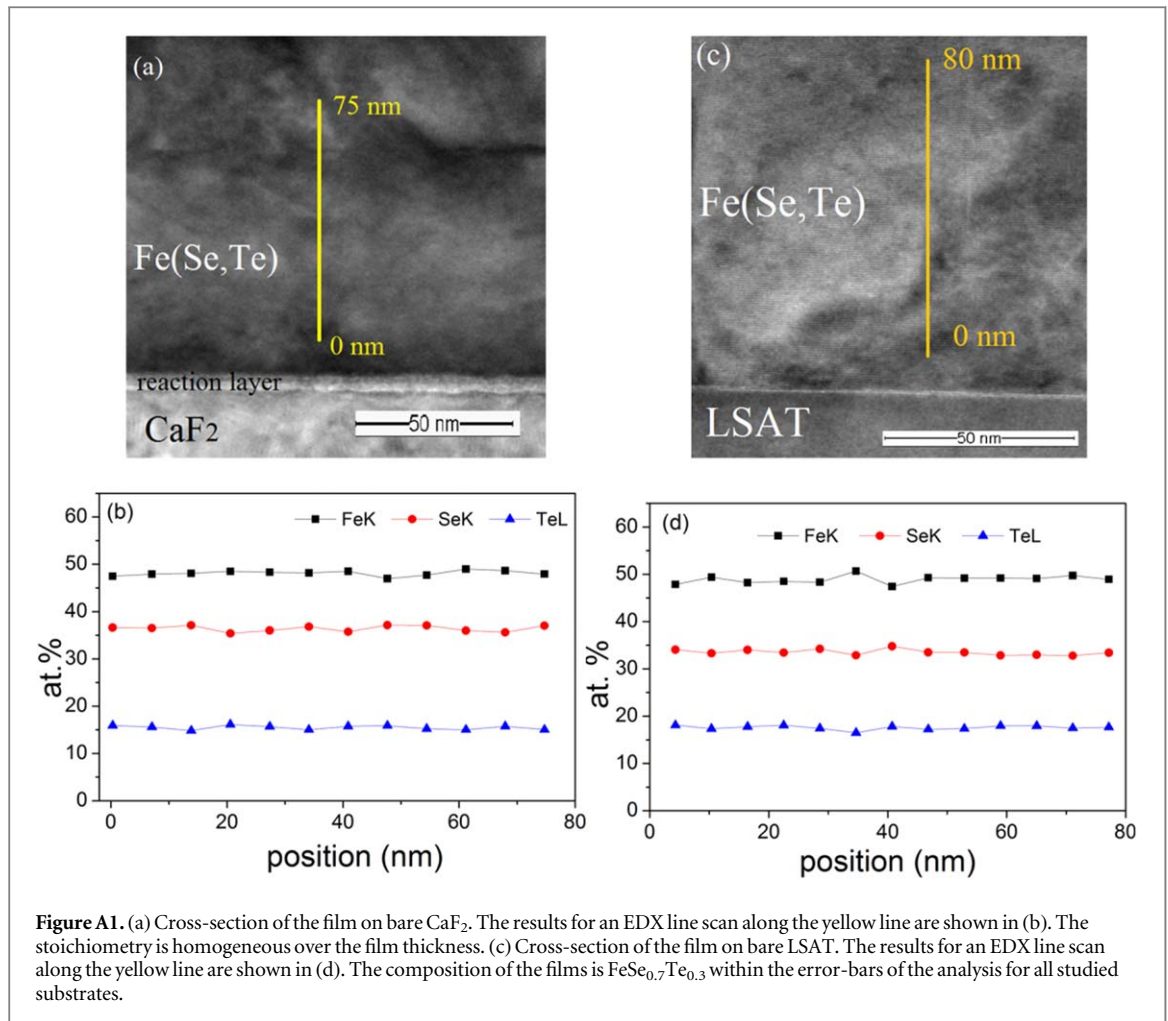


Figure A1. (a) Cross-section of the film on bare CaF₂. The results for an EDX line scan along the yellow line are shown in (b). The stoichiometry is homogeneous over the film thickness. (c) Cross-section of the film on bare LSAT. The results for an EDX line scan along the yellow line are shown in (d). The composition of the films is FeSe_{0.7}Te_{0.3} within the error-bars of the analysis for all studied substrates.

Acknowledgments

The authors thank S-L Drechsler, D Efremov and A Maeda for fruitful discussions and M Kühnel, U Besold for technical support. The research leading to these results has received funding from the National Science Foundation of China (Grant No. NSFC-U1432135, 11674054 and 11611140101) and Open Partnership Joint Projects of JSPS Bilateral Joint Research Projects (Grant No. 2716G8251b), the JSPS Grant-in-Aid for Scientific Research (B) Grant Number 16H04646 and the DFG funded GRK1621. VG is grateful to the DFG (GR 4667) for financial support. The publication of this article was funded by the Open Access Fund of the Leibniz Association.

Appendix

A.1. Structural properties

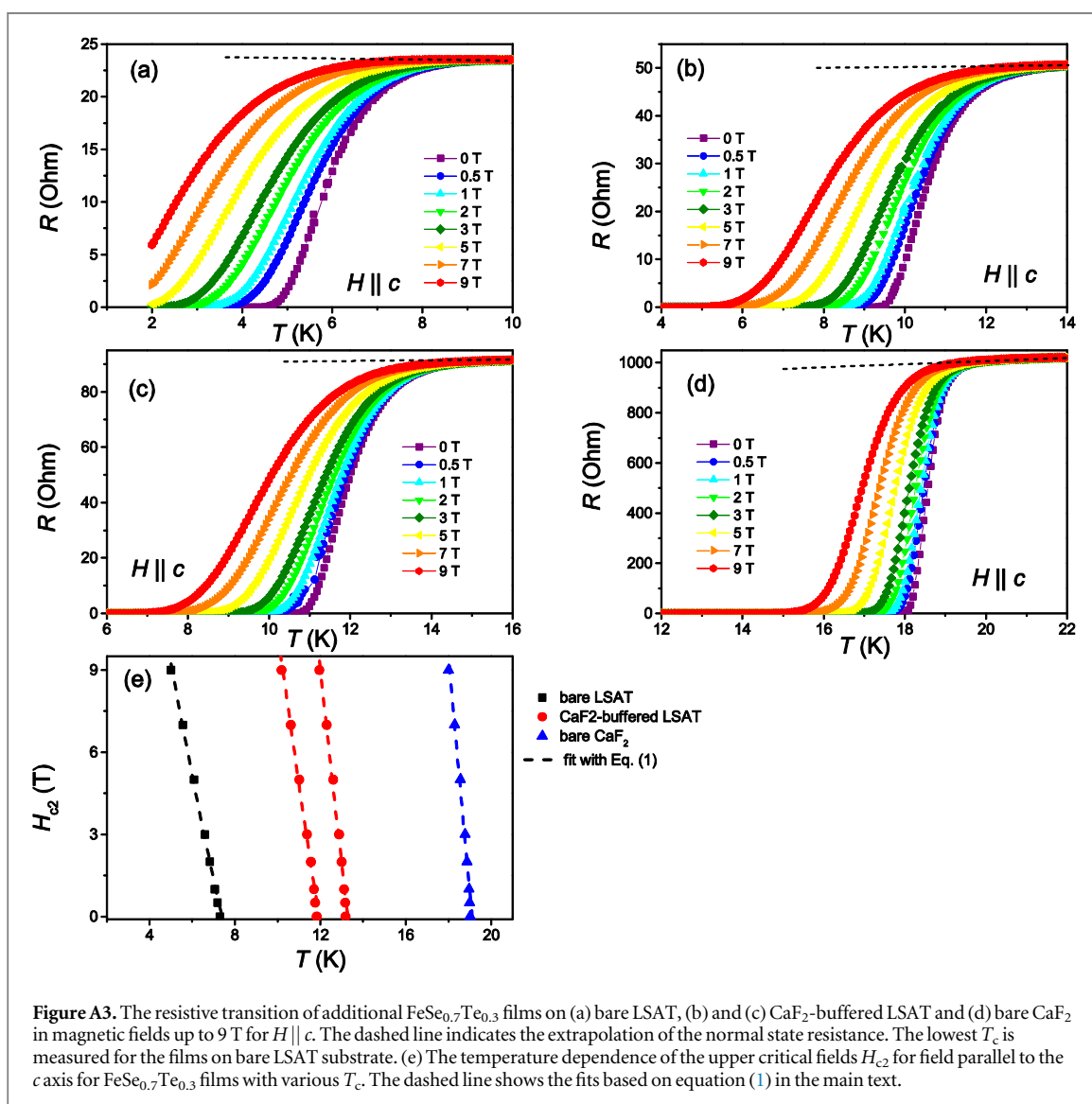
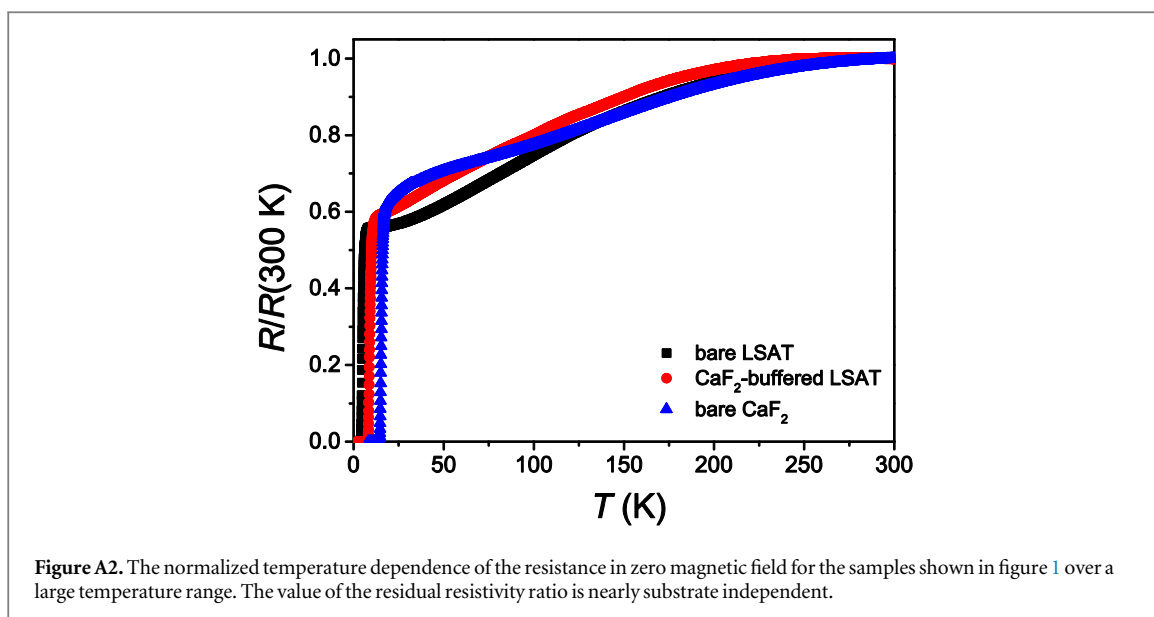
The structural properties and composition of the films were analyzed using TEM. The data for the two representative films are shown in figure A1 indicating a homogeneous stoichiometry over the film thickness.

A.2. Electrical resistance

In this section, we provide additional electrical resistivity data (not shown in the main text) measured in zero and applied magnetic field.

A.3. H_{c2} analysis

To take into account paramagnetic pair-breaking effects we used an analysis based on the Ginzburg–Landau theory, which provides a simple analytical expression for the temperature dependence of $H_{c2}(T)$ near T_c including paramagnetic effects [43]. As shown by Mineev *et al* H_{c2} can be calculated for clean single-band superconductors using the following equation:



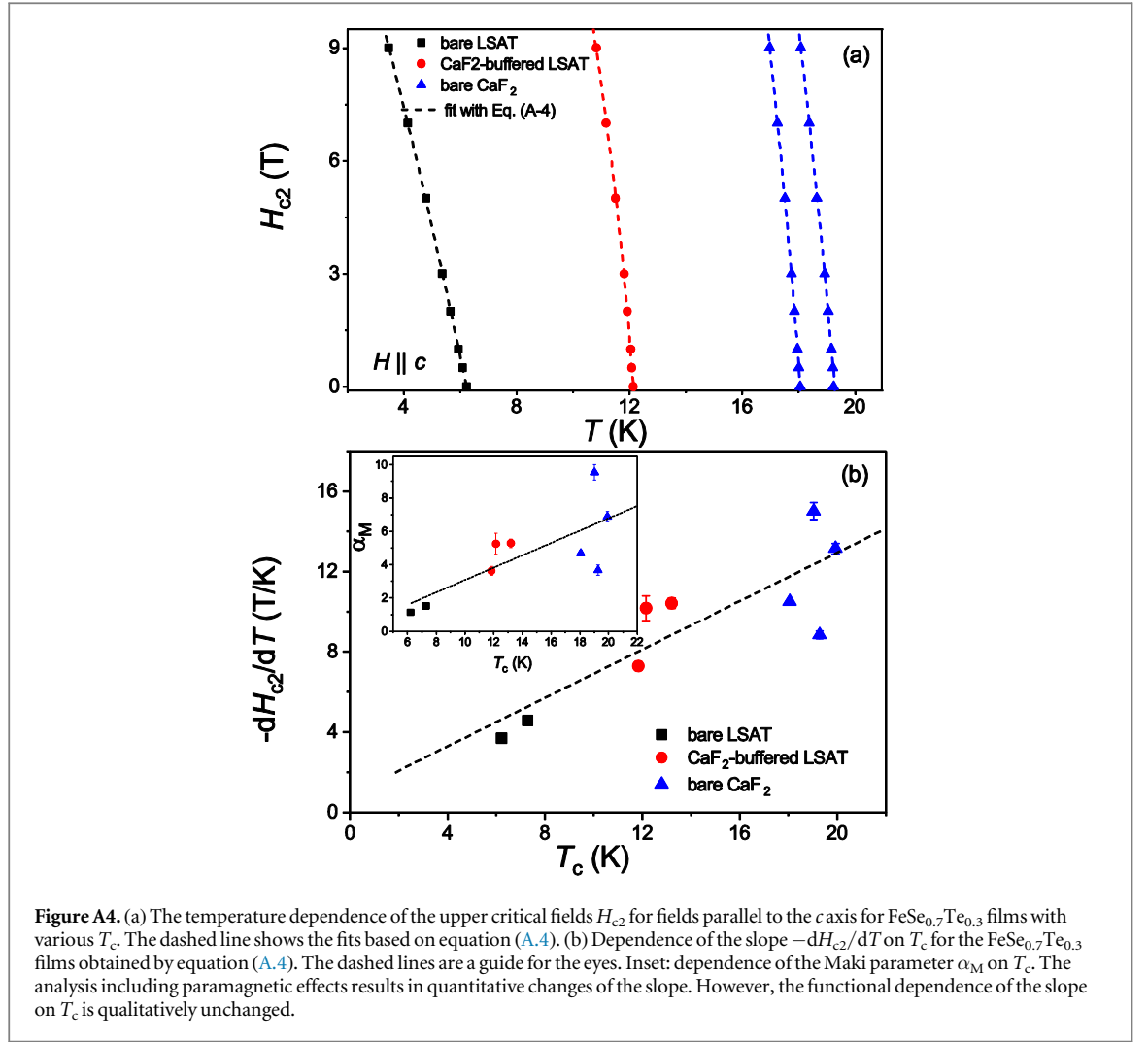


Figure A4. (a) The temperature dependence of the upper critical fields H_{c2} for fields parallel to the c axis for $\text{FeSe}_{0.7}\text{Te}_{0.3}$ films with various T_c . The dashed line shows the fits based on equation (A.4). (b) Dependence of the slope $-dH_{c2}/dT$ on T_c for the $\text{FeSe}_{0.7}\text{Te}_{0.3}$ films obtained by equation (A.4). The dashed lines are a guide for the eyes. Inset: dependence of the Maki parameter α_M on T_c . The analysis including paramagnetic effects results in quantitative changes of the slope. However, the functional dependence of the slope on T_c is qualitatively unchanged.

$$H_{c2} = \frac{e\gamma T_c^2}{a\mu^2} \left[-1 + \sqrt{1 + \frac{\alpha_0 a \mu^2}{(e\gamma T_c)^2} \frac{T_c - T}{T_c}} \right], \quad (\text{A.1})$$

where T_c is the critical temperature at zero field and $\alpha_0 = N_0$, $a = 7\zeta(3)N_0/4\pi^2$, $\gamma = 7\zeta(3)N_0v_F^2/32\pi^2T_c^2$. Here, N_0 is the density of states at the Fermi level, v_F is the Fermi velocity and μ is the magnetic moment.

The slope $-dH_{c2}/dT$ at T_c is given by:

$$-\frac{dH_{c2}}{dT}\Big|_{T_c} = \frac{\alpha_0}{2e\gamma T_c} \quad (\text{A.2})$$

with the Maki parameter defined as:

$$\alpha_M^2 = \frac{\alpha_0 a \mu^2}{(e\gamma T_c)^2}. \quad (\text{A.3})$$

Substituting equations (A.2) and (A.3) to equation (A.1) we obtain

$$H_{c2} = \frac{2}{\alpha_M^2} \left(-\frac{dH_{c2}}{dT}\Big|_{T_c} \right) T_c \left[-1 + \sqrt{1 + \alpha_M^2 \frac{T_c - T}{T_c}} \right]. \quad (\text{A.4})$$

We fitted our upper critical field data using equation (A.4), as shown by the dash line in figure A4. The obtained slope of the upper critical field $-dH_{c2}/dT$ is shown in figure A4 and the Maki parameter in the inset. Both quantities are proportional to T_c as expected (see also main text) [39].

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