In 2008, Kamihara et al.1 first discovered the iron-based superconductor LaO1−xFxFeAs, which has a superconducting critical temperature of 26 K. Subsequently, Hsu et al.2 reported that the binary superconductor FeSe with antifluorite planes has the transition temperature of 8 K. The applied pressure on the samples, the transition temperature can reach ~ 37 K.3,4 Ge et al.5 reported a superconducting transition temperature above 100 K in single-layer FeSe film grown on Nb-doped SrTiO3 (STO) substrate by molecular beam epitaxy method. Due to its simple crystal structure, this binary FeSe system with higher Tc is available, which has attracted tremendous interest in exploring the mechanism of high-temperature superconductivity.6–8 Generally, the FeSe layer is responsible for the superconductivity and the paired electrons are mainly 3d electrons of Fe ions. Meanwhile, the FeSe layers exhibit electrical neutrality, and the atoms between the layers are bonded together by van der Waals9,10. However, the same structure as FeTe does not show superconducting behavior. Yeh et al.11 found that when Te atoms are replaced by partially substituted Se atoms, the antiferromagnetic can be suppressed and its superconductivity is induced with a superconducting transition temperature of 15 K. In bulk crystals, the optimal Te content to achieve the highest superconductivity is 0.13–0.15 and phase separation occurs in the region of 0.1 < x < 0.2, whereas Tc increases with decreasing x for 0.2 ≤ x < 0.114. The interface effect between film and substrate makes it possible to obtain the FeSe1−xTe x films with high transition temperature in a metastable phase. Although researchers have done many studies on superconducting mechanism of FeSeTe films that prepared by pulsed laser deposition technology, there is a significant composition deviation between the nominal target and the thin film. Te doping can affect the Se/Te ratio and Fe content in chemical composition. The superconducting transition temperature of Fe0.76Se0.87Te0.13 and Fe0.91Se1−xTe x films is as high as 23 K at x = 0.6, and a sudden suppression of Tc is observed at 0.1 < x < 0.2, whereas Tc increases with decreasing x for 0.2 ≤ x < 0.115. The interface effect between film and substrate makes it possible to obtain the FeSe1−xTe x films with high transition temperature in a metastable phase. Although researchers have done many studies on superconducting mechanism of FeSeTe films that prepared by pulsed laser deposition technology (PLD), the bilayered effect of chemical composition on the superconductivity of FeSe1−xTe x films is uncertain.16–18 In this paper, we have prepared polycrystalline targets with different nominal composition to grow FeSe1−xTe x films and did a detailed investigation on the superconducting properties and its phase diagram. The experimental results show that there is a significant deviation between the nominal composition of targets and the real composition of films. The increase of Te doping can have an impact not only on Se/Te ratio but also on Fe content. The electrical transport results indicate that the optimal range of Te and Fe content is x = 0.13–0.15 and y = 0.73–0.78 for FeSe1−xTe x films with excellent superconductivity. As x = 0.13, y = 0.76, the maximum of...
Our previous work confirmed that TiO₂ as a buffer layer could increase the lattice match between Fe(Se, Te) films. We find that with Te doping, the (001) peak is observed along the c-axis for Fe(Se, Te) films. For 0.03 ≤ x ≤ 0.23 in Fig. 2a, as the temperature above the superconducting transition, the films only display metallic behavior. However, for x > 0.23, the resistivity of films changes from semiconducting to metallic before the superconducting transition. This change may attribute to the structural phase transition and magnetic phase transition caused by Te doping. If we define the point of intersection of the two lines as the normal-state resistivity ρ₀, the results are shown in Table 1. From Table 1, we think that the additional Fe may be incorporated in the inter-layer of Fe-Se/Te phase. Thus, Fe content plays a part in the change of lattice parameter. Zhuang et al. assumed that two key factors affected the lattice parameters of thin films under the Fe-deficient conditions. The above results show that the optimal chemical composition may play an important role in films with the excellent superconducting property. Based on this result, we measured the superconducting properties of these films and gave them in the following text. To explore the effect of Fe content on the superconductivity of Fe₅₋ₓSeₓTεₓ targets, we fixed the Se/Te ratio and change the Fe doping in the nominal composition, as shown in Table 2. It can be seen that the change of Fe doping in the nominal composition also affects the Fe content in the real composition, which may result in the obvious deviation of chemical composition between target and film. Therefore, we think that it is inaccurate to directly define the nominal composition of the targets as the real composition of the films. The semilogarithmic XRD patterns of Fe₅₋ₓSeₓTεₓ films are shown in Fig. 1. From Fig. 1, only Fe₅₋ₓSeₓTεₓ and TiO₂ peaks are observed along the c-axis (001), which indicates the Fe₅₋ₓSeₓTεₓ films to be the single tetragonal phase. Our previous work confirmed that TiO₂ as a buffer layer could increase the lattice match between Fe(Se, Te) film and STO substrate, so as to enhance the superconducting property of Fe(Se, Te) film. We find that with increasing Te doping, the (001) peaks significantly shift to a low angle. The c-axis lattice parameters for Fe₅₋ₓSeₓTεₓ films are obtained by fitting the (001) peak, as listed in Table 1. The ionic radius of Te (Te²⁻, 221 pm) is larger than that of Se (Se²⁻, 198 pm) which increases the c-axis lattice parameters. Zhuang et al. and Imai et al. have reported the effect of chemical composition on the structure in Fe₅₋ₓSeₓTεₓ films. In our results, the increase of Te doping in targets can also raise the Fe content in Fe₅₋ₓSeₓTεₓ films. For y > 1 in Table 1, we think that the additional Fe may be incorporated in the inter-layer of Fe-Se/Te phase. The Fe ionic radius is larger than that of Se and Te. Fe vacancy phase leads to a smaller c-axis lattice parameter, while Se/Te interstitial phase leads to a larger c-axis lattice parameter.

Table 1. The composition, onset and zero-resistivity temperature, and c-axis lattice parameter of thin films for nominal composition FeSe₀.₈₅Te₀.₁₅ targets.

| Nominal composition | Real composition (± 0.02) | Tc onset (K) | Te onset (K) | c parameter (Å) |
|---------------------|---------------------------|-------------|-------------|-----------------|
| Fe₁₀Se₄Te₄       | Fe₁₀Se₄Te₄               | 20.35       | 17.55       | 5.7287          |
| Fe₇Se₆Te₆       | Fe₇Se₆Te₆               | 18.95       | 17.34       | 5.8398          |
| Fe₅Se₈Te₈       | Fe₅Se₈Te₈               | 17.64       | 16.01       | 6.0047          |

Table 2. The composition, onset and zero-resistivity temperature, and c-axis lattice parameter of thin films for nominal composition Fe₅₋ₓSeₓTεₓ targets.

| Nominal composition | Real composition (± 0.02) | Tc onset (K) | Te onset (K) | c parameter (Å) |
|---------------------|---------------------------|-------------|-------------|-----------------|
| Fe₁₀Se₄Te₄       | Fe₁₀Se₄Te₄               | 20.35       | 17.55       | 5.7287          |
| Fe₇Se₆Te₆       | Fe₇Se₆Te₆               | 18.95       | 17.34       | 5.8398          |
| Fe₅Se₈Te₈       | Fe₅Se₈Te₈               | 17.64       | 16.01       | 6.0047          |

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The values of Tc and Tρ for these films are listed in Table 1 and plotted in the 3D phase diagram, as shown in Fig. 2c. With increasing the Te doping, the Tc rises at first and then decreases. From Fig. 2c, the Fe0.76Se0.87Te0.13 film exhibits the higher Tc and Tρ about 18.95 K and 17.34 K, respectively. Surprising us, the composition

Fe0.76Se0.87Te0.13 film is not consistent with that of the single crystal, where the highest Tc is considered x ≈ 0.6 in Fe(Se1−xTe0.3) polycrystal sample, and located at the phase separation region of 0.1 ≤ x ≤ 0.312. They argued that the single-phase of Fe(Se1−xTe0.3) single crystals with the region of 0.1 ≤ x ≤ 0.3 were not easy to obtain. However, Imai et al.14 assumed that the single-phase epitaxial films of FeSe1−xTe0.1 with 0.1 ≤ x ≤ 0.4 could be successfully prepared on CaF2 substrates, attributing to the strain effect between film and substrate. Due to the different substrates, there is a difference in the suppression of phase separation and giant enhancement of Tc for FeSe1−xTe0.1 films. Our experimental results display that the sudden suppression of Tc is observed at 0.03 ≤ x < 0.13, whereas Tc increases with decreasing x for 0.13 ≤ x ≤ 0.56. The superconductivity is related to the Te and Fe content in FeSe1−yTe0.1 films. Therefore, we must consider the effects of Fe vacancies on the superconductivity of FeSe1−yTe0.1 films. Figure 2b shows the temperature dependence of the normalized resistivity ρ/ρ0(ρ-T) near the optimal composition FeSe1−yTe0.1 films, where x = 0.15 and y = 0.76. The results demonstrate the effects of Fe vacancies on the superconductivity of FeSe1−yTe0.1 films. The Tc onset and Tρ are listed in Table 2 and plotted in the 3D phase diagram of Fig. 2b. Although we do not know why the Tc onset and Tρ increase with decreasing the Fe content near y = 0.76, the transition width broadens much more. This result further confirms that the optimal range is x = 0.13–0.15 and y = 0.73–0.78 for the FeSe1−yTe0.1 films.

Figure 2c is a new 3D phase diagram for the FeSe1−yTe0.1 films. The blue open symbols are the projection of experimental points on the xy-plane at Tc ≈ 1 K. The 3D phase diagram can be divided into three regions by Tρ onset(x, y) and Tc(x, y) curved surfaces, which are superconductivity (SC), flux flow (FF), and normal state (NS), respectively. Above the Tρ onset(x, y) curved surfaces, the FeSe1−yTe0.1 film is in the normal state. Below the Tc(x, y) curved surfaces, the FeSe1−yTe0.1 film is in a superconducting state. Between the Tρ onset(x, y) and Tc(x, y) curved surfaces, the FeSe1−yTe0.1 film is in the flux flow state. The 3D phase diagram demonstrates that the phase separation is absent, and that the optimal composition for the FeSe1−yTe0.1 film on STO substrate is not x ≈ 0.5 and y = 1 but x = 0.13 and y = 0.76. It should be noted that the dependence of Tc on x suddenly changes at the boundary defined by 0.03 ≤ x < 0.13 in our experiment. Thus, not only the decrease of Tc with x ≥ 0.13 can be explained by the empirical law that shows the relation between Tc and structural parameters, but also the sudden suppression of Tc in films with 0.03 ≤ x < 0.13 can be explained by the orthorhombic distortion results in a suppression of Tc. As reported by Imai et al.14, the orthorhombic distortion is applicable to the behavior of films, if a large orthorhombic distortion is observed only in films with 0 < x < 0.1, which is consistent with our result of 0.03 ≤ x < 0.13. Chen et al.28 and Bendele et al.29 pointed out that a few Fe vacancies were beneficial to improve the superconductivity and raised the superconducting transition temperature for FeSe1−yTe0.1 films. The inhomogeneous distribution of Fe vacancies can induce the Fe disorder effect in FeSe1−yTe0.1 films with x < 1. The first-principles calculation also showed that the Fe vacancies could effectively increase the number of electron carriers and change the electronic properties in samples.28 Therefore, in this experiment, the highest Tc onset and Tρ occur near y = 0.76. When the Te and Fe content exceed the optimal composition, the Tc onset' and the Tρ' of FeSe1−yTe0.1 films decrease. For example, as x = 0.56, y = 1.43, the ρ does not down to 1% ρ0, so the Fe1.43Se0.56Te0.56 film only has the Tc onset' about 8.03 K.

To understand the new phase diagram, we have measured the electrical transport and magnetization properties for FeSe1−yTe0.1 films in magnetic field. Here, we choose some typical results in the next part. Figure 3a,b present the temperature dependence of resistivity of Fe0.76Se0.87Te0.13 film in various magnetic fields up to 9 T applied perpendicular and parallel to the c-axis. With increasing the applied magnetic field, the resistive transition

Figure 1. Semilogarithmic X-ray diffraction patterns of FeSe1−yTe0.1 thin films.
is broadened. At the same field, the width of superconducting transition $\Delta T_c$ for $H_{//c}$ is larger than that for $H_{//ab}$. This result indicates that the Fe$_y$Se$_{1-x}$Te$_x$ films are anisotropic near $T_c$.

If we define the onset transition temperature $T_{c,onset}$ as the critical temperature $T_c$, namely the field is the upper critical field $H_{c2}$, we can get the temperature of the upper critical field near $T_c$. The $H$-$T$ phase diagram for Fe$_{0.72}$Se$_{0.94}$Te$_{0.06}$, Fe$_{0.76}$Se$_{0.87}$Te$_{0.13}$ and Fe$_{0.91}$Se$_{0.77}$Te$_{0.23}$ films is shown in Fig. 4a. The temperature dependence of the upper critical field $H_{c2}$ near $T_c$ follows the formula $H_{c2}(T) = H_{c2}(0)(1 - T/T_c)^n$, where $H_{c2}(0)$ and $n$ are parameters obtained from the experimental data. The parameters $H_{c2}(0)$ and $n$ for $H_{c2}$ (0) are parameters obtained from the experimental data. The parameters $H_{c2}(0)$ and $n$ for $H_{c2}$ and $H_{c2}$ near $T_c$ for (1) Fe$_{0.72}$Se$_{0.94}$Te$_{0.06}$, (2) Fe$_{0.76}$Se$_{0.87}$Te$_{0.13}$ and (3) Fe$_{0.91}$Se$_{0.77}$Te$_{0.23}$ films, respectively, are (1) 67.9 T and 0.63 for $H_{//ab}$, 46.6 T and 0.78 for $H_{//c}$; (2) 91.8 T and 0.54 for $H_{//ab}$, 82.5 T and 0.71 for $H_{//c}$; and (3) 77.4 T and 0.71 for $H_{//ab}$, 60.2 T and 0.53 for $H_{//c}$. The result implies that the upper critical field $H_{c2}$ depends on Te and Fe content. The higher upper critical field $H_{c2}$ located at $x = 0.13-0.15$ and $y = 0.73-0.78$ for Fe$_{0.72}$Se$_{0.94}$Te$_{0.06}$ films. From Fig. 4a, we can get the temperature dependence of the anisotropic factor $\gamma = H_{c2}(H_{//ab})/H_{c2}(H_{//c})$ near $T_c$, as shown in Fig. 4b. We can see that the $\gamma$ value decreases with decreasing temperature. $\gamma$ for Fe$_{0.72}$Se$_{0.94}$Te$_{0.06}$, Fe$_{0.76}$Se$_{0.87}$Te$_{0.13}$ and Fe$_{0.91}$Se$_{0.77}$Te$_{0.23}$ films are estimated about 3.3, 1.9 and 1.6, respectively. Increasing Te doping can inhibit its anisotropy and enhance the correlation between Fe-Se/Te layers, leading to the increasing the dimensionality of Fermi surface, which is conducive to the transmission of electrons along the c-axis direction.

Figure 2. (a,b) Temperature dependence of resistivity from 2 to 300 K for Fe$_x$Se$_{1-x}$Te$_x$ thin films. (a) $(x, y) = (0.03, 0.63), (0.06, 0.72), (0.13, 0.76), (0.23, 0.91), (0.34, 1.09)$ and $(0.56, 1.43)$. Inset: enlarged plot for the definition of normal-state resistivity $\rho_n$. (b) $(x, y) = (0.15, 0.73), (0.13, 0.76)$ and $(0.16, 0.78)$. Inset: the enlarged $\rho$-$T$ curve near $T_c$. (c) Sketch of the proposed temperature doping 3D phase diagram for Fe$_x$Se$_{1-x}$Te$_x$ superconducting system, showing regions of superconductivity (SC), flux flow (FF) and normal state (NS).
The effective pinning energy is an important parameter to enhance the capacity of carrying current for superconducting materials. According to the thermally activated flux flow (TAFF) theory, the $\ln \rho - 1/T$ in the TAFF region can be described using an Arrhenius relation$^{30}$, 

$$\rho = \rho_0 \exp \left( - \frac{U_0}{k_B T} \right)$$

where $U_0$ is the effective pinning energy. Figure 5a,b shows the linear relationship between $\ln \rho$ and $1/T$ of the Fe$_{0.76}$Se$_{0.87}$Te$_{0.13}$ film. From the absolute slope of $\ln \rho - 1/T$ curves, we can obtain the effective pinning energy $U_0$ of Fe$_{0.72}$Se$_{0.94}$Te$_{0.06}$, Fe$_{0.76}$Se$_{0.87}$Te$_{0.13}$, and Fe$_{0.91}$Se$_{0.77}$Te$_{0.23}$ films, respectively, as shown in Fig. 5c. It can be found that the $U_0$ value of Fe$_{0.76}$Se$_{0.87}$Te$_{0.13}$ is larger than that of Fe$_{0.72}$Se$_{0.94}$Te$_{0.06}$ and Fe$_{0.91}$Se$_{0.77}$Te$_{0.23}$ in the same field. What's more, $U_0$ values for $H//ab$ plane are much higher than that for $H//c$, indicating the flux pinning is anisotropic. The magnetic field dependence of $U_0$ follows a power law $U_0(H) \sim H^{-\alpha}$. When $H//ab$, the parameter $\alpha$ decreases with increasing the Te doping. The parameter $\alpha$ for the Fe$_{0.76}$Se$_{0.87}$Te$_{0.13}$ and Fe$_{0.91}$Se$_{0.77}$Te$_{0.23}$ films is close. For $H//c$, there is an obvious crossover occurred at $H \approx 2$ T. Below 2 T, the parameter $\alpha$ is close to 0.15. Above 2 T, $\alpha$ is close to 0.5. Similar behavior has been observed in other superconductors$^{30-34}$. In the field below 2 T, the pinning energy $U_0$ is weakly dependent on the applied magnetic field $H$. It can be considered that the number of magnetic flux lines is much less than the number of pinning centers. The single vortex pinning dominates in this region$^{35}$. As the magnetic field increases above 2 T, more flux lines enter the superconductor and the flux spacing becomes smaller, which leads to the pinning energy being inhibited. The pinning energy $U_0$ becomes strongly dependent on the field $H$. and the collective creep pinning is dominant in this region$^{36,37}$.

Figure 3. Temperature dependence of resistivity near $T_c$ in various magnetic fields for Fe$_{0.76}$Se$_{0.87}$Te$_{0.13}$ thin film. (a) $H//c$ and (b) $H//ab$.
The critical current density $J_c$ is also an important parameter for high quality epitaxial superconducting films. To study the effect of chemical composition on the critical current density of $\text{Fe}_y\text{Se}_{1-x}\text{Te}_x$ films, we have measured the magnetization hysteresis loops in fields parallel to the $c$-axis from 0 to ±9 T. Figure 6 shows the $M$-$H$ loops of $\text{Fe}_{0.91}\text{Se}_{0.77}\text{Te}_{0.23}$ film at various temperatures. The $M$-$H$ loops show symmetric field dependence. As the field increases, the magnetization of film decreases. The critical current density $J_c$ is estimated from the $M$-$H$ loops by the Bean critical state model:

$$J_c = \frac{2\mu_0 \Delta M}{a(1-a/3b)}.$$  

Where $\Delta M = M(+) - M(-)$, $M(+) and M(-)$ are the magnetizations when sweeping fields up and down, respectively. $a$ and $b (a < b)$ are the $\text{Fe}_y\text{Se}_{1-x}\text{Te}_x$ film’s cross-sectional dimension. The field dependence of the critical current density $J_c$ at various temperatures is shown in Fig. 7. From Fig. 7a–c, we can see that with increasing the Te doping, the field dependence of the critical current density $J_c$ improves at higher field region. In addition, the measured critical temperature $T_c$ in $\text{Fe}_{0.76}\text{Se}_{0.87}\text{Te}_{0.13}$ is higher than that in $\text{Fe}_{0.91}\text{Se}_{0.77}\text{Te}_{0.23}$. The calculated $J_c$ at 4 K and 0 T for $\text{Fe}_{0.72}\text{Se}_{0.94}\text{Te}_{0.06}$, $\text{Fe}_{0.76}\text{Se}_{0.87}\text{Te}_{0.13}$, $\text{Fe}_{0.91}\text{Se}_{0.77}\text{Te}_{0.23}$ films are about $4.46 \times 10^5$ A/cm², $4.51 \times 10^6$ A/cm² and $4.05 \times 10^6$ A/cm², respectively. This result displays that the higher $T_c$ also contributes to improving the magnetic field dependence of $J_c$ at 4 K. Therefore, the optimal composition is beneficial for $\text{Fe}_y\text{Se}_{1-x}\text{Te}_x$ films exhibiting excellent superconductivity in lower field region.

Flux pinning force can provide a very efficient route to describe the vortex dynamics in superconductors. Furthermore, we calculated the field dependence of the flux pinning force $F_p = \mu_0 H \times J_c$ for temperatures at 11, 12 and 13 K, respectively. Based on the theory of Dew-Hughes, the field dependence of the normalized vortex pinning force $f_p$ should follow the expression $f_p = Ah^q(1 - h)^p$, where $h = H/H_{c2}$, $p$ and $q$ are parameters that depend on the pinning centers. Figure 7d gives the relationship of normalized vortex pinning force $f_p$ and reduced magnetic field $h$ for $\text{Fe}_{0.76}\text{Se}_{0.87}\text{Te}_{0.13}$ film. By fitting the $f_p$-$h$ curves, we obtain $p = 0.67$, $q = 2.45$, and $h_{max} = 0.21$, indicating that the flux pinning centers in film may be dominant by the core normal surface pinning ($p=0.5$, $q=2$, and $h_{max} = 0.2$).
The interface structure plays a vital role in determining the superconducting properties for Fe$_{y}$Se$_{1-x}$Te$_{x}$ films. Using the STEM analysis, we could reveal the Fe$_{0.76}$Se$_{0.87}$Te$_{0.13}$/TiO$_2$/STO microstructure and determine the morphology of the interface. The thicknesses of Fe$_{0.76}$Se$_{0.87}$Te$_{0.13}$ and TiO$_2$ film are about 32.4 nm and 29.5 nm, respectively. Figure 8a shows the overview image of the Fe$_{0.76}$Se$_{0.87}$Te$_{0.13}$/TiO$_2$/STO interface. It can be seen that the heterostructure interface is sharp and clean. The TiO$_2$ buffer was successfully deposited between the Fe$_{0.76}$Se$_{0.87}$Te$_{0.13}$ film and STO substrate. Figure 8b shows the high-magnification HAADF image of Fe$_{0.76}$Se$_{0.87}$Te$_{0.13}$/TiO$_2$.

Figure 5. lnρ versus 1/T curves in various magnetic fields of Fe$_{0.76}$Se$_{0.87}$Te$_{0.13}$ thin film. (a) H//c; (b) H//ab. (c) Magnetic field dependence of the effective flux pinning energy for Fe$_{0.72}$Se$_{0.94}$Te$_{0.06}$, Fe$_{0.76}$Se$_{0.87}$Te$_{0.13}$ and Fe$_{0.91}$Se$_{0.77}$Te$_{0.23}$ thin films.

Figure 6. Magnetic hysteresis loops of Fe$_{0.91}$Se$_{0.77}$Te$_{0.23}$ thin film at various temperatures in magnetic field parallel to the c-axis.

The interface structure plays a vital role in determining the superconducting properties for Fe$_y$Se$_{1-x}$Te$_x$ films. Using the STEM analysis, we could reveal the Fe$_{0.76}$Se$_{0.87}$Te$_{0.13}$/TiO$_2$/STO microstructure and determine the morphology of the interface. The thicknesses of Fe$_{0.76}$Se$_{0.87}$Te$_{0.13}$ and TiO$_2$ film are about 32.4 nm and 29.5 nm, respectively. Figure 8a shows the overview image of the Fe$_{0.76}$Se$_{0.87}$Te$_{0.13}$/TiO$_2$/STO interface. It can be seen that the heterostructure interface is sharp and clean. The TiO$_2$ buffer was successfully deposited between the Fe$_{0.76}$Se$_{0.87}$Te$_{0.13}$ film and STO substrate. Figure 8b shows the high-magnification HAADF image of Fe$_{0.76}$Se$_{0.87}$Te$_{0.13}$/TiO$_2$. The
Fe, Se, Te, Ti and O atoms are arranged neatly at the interface. In this case, the \( \text{Fe}_{0.76}\text{Se}_{0.87}\text{Te}_{0.13} \) structure with a tetragonal space group \( P4/nmm \) is very simple, and each unit cell contains 3 quintuple layers (QLs), which are bonded by van der Waals (vdW)\(^9\). The \( \text{TiO}_2 \) unit cell has two Ti–O triple layers, which grow on STO along the (00\(l\)) direction. From Fig. 8b, a nanoscale damaged layer (or transition layer) was formed between the \( \text{TiO}_2 \) and \( \text{Fe}_{0.76}\text{Se}_{0.87}\text{Te}_{0.13} \) interface. To determine the formation of this transition layer, the Atomic resolution EDX mapping was conducted in this area. The chemical elemental maps of Fig. 8c confirm the suggestion from HAADF image that the atoms are arranged regularly without obvious diffusion and migration. Such high quality heterostructure has a significant influence on the enhancement of superconductivity for \( \text{Fe}_{y}\text{Se}_{1-x}\text{Te}_x \) films.

**Conclusion**

In summary, we successfully prepared the \( \text{Fe}_{y}\text{Se}_{1-x}\text{Te}_x \) thin films with \( 0.03 \leq x \leq 0.56 \) and \( 0.63 \leq y \leq 1.43 \) by PLD. Our experimental results confirm the significant deviation between the nominal compositions of targets and the real compositions of \( \text{Fe}_{y}\text{Se}_{1-x}\text{Te}_x \) films. Chemical composition does affect the superconducting properties such as the superconducting transition temperature and the critical current density in \( \text{Fe}_{y}\text{Se}_{1-x}\text{Te}_x \) films. A new 3D phase diagram is presented from the experimental results of electrical transport, which reveals that the optimal composition for \( \text{Fe}_{y}\text{Se}_{1-x}\text{Te}_x \) films is \( x = 0.13–0.15 \) and \( y = 0.73–0.78 \). The field dependence of flux pinning energy displays that the increase of Te doping can enhance the flux pinning in \( \text{Fe}_{y}\text{Se}_{1-x}\text{Te}_x \) films. STEM investigation shows that the \( \text{Fe}_{y}\text{Se}_{1-x}\text{Te}_x /\text{TiO}_2/\text{STO} \) heterostructure has a sharp interface and exhibits almost no atomics intermixing. Our study results provide some further understanding on the mechanism of superconducting properties for \( \text{Fe}_{y}\text{Se}_{1-x}\text{Te}_x \) films, which has a certain guiding significance and reference value for the potential application of iron-based superconductors.
Methods

The PLD targets were prepared by the self-flux method with high purity materials (Fe 99.99%, Te 99.999% and Se 99.999%) in the stoichiometric proportion. Fe, Se and Te were fully ground and squeezed into a 3/4 in. block, and then encapsulated in a vacuum quartz tube. The vacuum quartz tube was calcined in a muffle furnace at 850 °C for 72 h, then slowly cooled down to room temperature at the rate of 3 °C/min. The Fe\textsubscript{y}Se\textsubscript{1−x}Te\textsubscript{x} epitaxial films were deposited on STO single crystalline substrates at 300 °C by PLD in a high vacuum (~ 10⁻⁷ mbar). The distance between target and substrate was set at ~ 70 mm. A KrF excimer laser (248 nm) was used for deposition with an energy density of 2.0 J/cm² and a repetition frequency of 2 Hz. The size of the STO substrate is 5 mm × 5 mm. TiO\textsubscript{2} film as a buffer layer was firstly deposited on STO substrate by PLD to improve the lattice matching between Fe\textsubscript{y}Se\textsubscript{1−x}Te\textsubscript{x} film and STO substrate. The deposition temperature and deposition time for Fe\textsubscript{y}Se\textsubscript{1−x}Te\textsubscript{x} and TiO\textsubscript{2} films were 300 °C and 15 min, 600 °C and 4.5 min, respectively. After deposition, the films were annealed to room temperature at the rate of 5 °C/min.

X-ray diffraction (XRD) patterns using the θ/2θ method were measured by Bruker D8 with CuKα radiation (λ = 1.54 Å). The Φ-scan of (101) peak from the Fe\textsubscript{0.76}Se\textsubscript{0.87}Te\textsubscript{0.13} thin film is shown in Supplementary SFig. 1. The chemical composition of Fe\textsubscript{y}Se\textsubscript{1−x}Te\textsubscript{x} films was determined by energy dispersive x-ray spectroscopy (EDX) in a Gemini 500 scanning electron microscope (SEM) mapping. The measurements of electrical transport were carried out via the physical property measurement system (PPMS-9T, Quantum Design). Magnetization measurements on films with 100 Oe/s of sweep rate were performed in vibrating sample magnetometer (VSM). The microstructures of Fe\textsubscript{y}Se\textsubscript{1−x}Te\textsubscript{x} films were examined by scanning transmission electron microscopy (STEM, FEI Titan G2 60–300 aberration). Samples for the STEM were cut and milled in a focused ion beam (FIB, FEI Helios Nanolab 600) according to the so-called micro-bridge sampling technique.

Figure 8. (a) Overview image of the Fe\textsubscript{0.76}Se\textsubscript{0.87}Te\textsubscript{0.13}/TiO\textsubscript{2}/STO thin film interface. (b) Atomically resolved HADDF-STEM image of Fe\textsubscript{0.76}Se\textsubscript{0.87}Te\textsubscript{0.13}/TiO\textsubscript{2} heterostructure. (c) EDX-mapping results shows the distribution of Fe (red), Se (green), Te (blue) Ti (magenta), O (cyan).
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Y.Z., T.W., Z.W. and Z.X. conceived the experiments; Y.Z. conducted the experiments; Y.Z., Z.W., and Z.X. analysed the results; Y.Z. and Z.W. wrote the paper. All authors have reviewed the manuscript.

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The authors declare no competing interests.

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