The vortex state of $\text{FeSe}_{1-x}\text{Te}_x$ superconducting thin films

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Abstract

We report the vortex dynamics of tellurium substituted $\text{FeSe}_{1-x}\text{Te}_x$ superconducting thin films. The electric field versus current density (E – J) curve for films with low Te substitution is still governed by the thermally activated flux flow model at temperatures as low as $0.5T_C^{\text{offset}}$. In contrast, we clearly observed a vortex liquid–glass transition in films with high Te substitution. The E – J curves of these samples fit nicely to the scaling relations based on the 3D vortex glass theory. Our results reveal an enhancement of the vortex pinning as more Te content is introduced, which probably originates from the excess Fe at the interstitial site.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The discovery of iron-based superconductors [1–3] has attracted enormous worldwide attention, similar to what followed the discovery of high T_C cuprate superconductors more than twenty years ago. In addition to providing the opportunity for achieving a better understanding of the superconductivity mechanism, the high upper critical field and low anisotropy indicate promise for applications [1–4]. Increasing the critical current density J_C is an important task for applications. The J_C of iron-based superconducting thin film was enhanced by introducing a buffer layer and extrinsic pinning centers [5, 6]. Superconducting quantum interference devices composed from grain boundary junctions were also achieved [7]. However to get better improvement of J_C , a more detailed understanding of the pinning mechanism and vortex state properties are necessary. Kim *et al* reported evidence of a 3D vortex glass state in $(\text{Ba}, \text{K})\text{Fe}_2\text{As}_2$ single crystal [8]. On the other hand, thermally activated flux flow was observed in $\text{NdFeAsO}_{0.75}\text{F}_{0.25}$ [9] and $\text{Fe}_{1.03}\text{Te}_{0.55}\text{Se}_{0.45}$ [10] crystals. Strong vortex pinning, with pinning centers due to spatial fluctuation of superconducting areas or including

excess Fe, was reported in $\text{Fe}_{1.04}\text{Te}_{0.60}\text{Se}_{0.40}$ single-crystalline samples [11]. The existence of a vortex liquid–glass transition in $\text{FeSe}_{1-x}\text{Te}_x$ superconductors is still not proved, and a more systematic study may help to better characterize the vortex state. In this paper, we report the vortex states of a series of $\text{FeSe}_{1-x}\text{Te}_x$ superconducting thin films. The detailed E – J characteristics reveal the difference in vortex behavior between low Te substitution and high substitution samples. A clear vortex glass–liquid transition is observed in high Te substitution samples. However even well below T_C^{offset} , thermally activated behavior still dominates the motion of the vortex in low Te substitution samples and no signature of a vortex glass–liquid transition appears.

2. Experimental details

The c -axis preferred orientation $\text{FeSe}_{1-x}\text{Te}_x$ thin films were grown on an MgO substrate by the pulsed laser deposition (PLD) technique. The details of the sample preparation and characterization were described earlier [12]. The Te substitution level of these thin films was calibrated on the

Figure 1. ((a)–(e)) The resistive transition of $\text{FeSe}_{1-x}\text{Te}_x$ films, $x = 0, 0.43, 0.48, 0.56$, and 0.85 , as a function of temperature. The applied magnetic fields are 0, 1, 3, 5, 7, 9 T along the c -axis of the film. (f) The magnetic field dependence of T_C^{offset} and ΔT_C . The superconducting transition becomes less broadened under magnetic fields in higher Te substitution samples, except for the $x = 0.85$ sample. The applied current density during measurement is around 2 kA cm^{-2} .

basis of the position of the x-ray diffraction peaks, and double-checked via the chemical composition determined using EDAX. The films were patterned into a Hall-bar geometry for transport measurements, which were performed using a Quantum Design physical property measurement system (QD-PPMS). The magnetic field was applied along the c -axis of the thin film.

3. Results and discussion

Figures 1(a)–(e) show the resistive transition for $\text{FeSe}_{1-x}\text{Te}_x$ films as a function of temperature under magnetic fields up to 9 T for $x = 0, 0.43, 0.48, 0.56$, and 0.85 respectively. The offset superconducting transition temperature, T_C^{offset} , defined as $\rho(T_C^{\text{offset}}) = 0.1\rho_n$, and the superconducting transition width, $\Delta T_C \equiv T(0.9\rho_n) - T(0.1\rho_n)$, are shown in figure 1(f). The superconducting transition temperature of $\text{FeSe}_{1-x}\text{Te}_x$ films is lower than that of polycrystalline samples, especially for low Te substitution films, because of the strain effect on thin films [13]. Under an external magnetic field, the superconducting transition shifts to low temperature and becomes broadened. In figure 1(f), the broadening of the superconducting transition is reduced in higher substitution samples ($x = 0.43, 0.48$, and 0.56), indicating a stronger flux pinning effect and a different vortex state at low temperature from low Te substitution films. A finite resistance at low

Figure 2. The ϕ -scan of (101) peaks from $\text{FeSe}_{1-x}\text{Te}_x$ films with $x = 0.56$ and 0.85 . In the $x = 0.56$ sample, two domains with fourfold symmetry can be observed, which are rotated by 45° with respect to each other. However, two extra shoulders were observed in the major domain peaks of $x = 0.85$ sample, indicating that two extra domains with different orientations exist in this particular sample.

temperature is observed in the $x = 0.85$ sample under high external magnetic fields (in figure 1(e)). The detailed in-plane x-ray diffraction results show that there may exist two different

Figure 3. ((a), (b)) The E - J curves of $\text{FeSe}_{1-x}\text{Te}_x$ films under magnetic fields of 3 T, for $x = 0.43$ and 0.48 film. The temperature range is from 7.75 to 4.2 K and from 8 to 4 K, respectively. The $\ln \rho$ - $1/T$ plot, shown in the inset, indicates free flux flow (FFF) and thermally activated flux flow (TAFF) regimes. No signature of a vortex glass transition was observed even at temperatures down to $0.5T_C^{\text{offset}}$ and $0.4T_C^{\text{offset}}$ for $x = 0.43$ and 0.48 films respectively.

grains with different orientations in this particular sample, as shown in figure 2. This finite resistivity at low temperature under strong magnetic field could be due to the suppression of the superconductivity of the grain boundaries.

Figure 3 displays a typical logarithmic plot of E - J curves at different temperatures for $x = 0.43$ and 0.48 samples under $B = 3$ T. For $x = 0.43$ sample, the temperature of these E - J curves ranges from 7.75 to 4.2 K, corresponding to the reduced temperature, defined as $T/T_C^{\text{offset}}(B = 0 \text{ T})$, ranging from 0.9 to 0.5. However even at $T/T_C^{\text{offset}}(B = 0 \text{ T}) = 0.5$, the curvatures of the E - J curves remain positive, revealing that the motion of the vortices is still within the thermally activated flux flow (TAFF) regime. Similar results were found for the $x = 0.48$ sample, as shown in figure 3(b), with $T/T_C^{\text{offset}}(B = 0 \text{ T})$ ranging from 0.8 to 0.4.

According to the TAFF model [14, 15], for the low J regime the E - J relation can be given as approximated as

$$E \approx 2\nu_0 \frac{JB^2V_cL_P}{k_B T} L \exp(-U_0/k_B T), \quad (1)$$

where U_0 is the activation energy for the hopping of vortex bundles, L_P is the pinning range, V_c is the flux bundle

volume and ν_0 is the attempt frequency. The Arrhenius plot, $\ln \rho$ versus $1/T$, is shown in the inset. Two slopes at low temperature are attributed to free flux flow and thermally activated flux flow regimes. The activation energy U_0 can be estimated from the slope of the Arrhenius plot for the TAFF regime. Figure 4 shows the magnetic field dependence of U_0 for $x = 0.43$ and 0.48 films. The U_0 is about a few hundred $k_B T$, which is smaller than those of Fe-based superconducting crystals [9, 10] and other superconductors [16, 17]. The U_0 follows a form $U_0 = \alpha(1 - B/B^*)^2$, where B^* is the irreversibility field B_{irr} or a Kramer scaling field B_K [17] (solid lines in figure 4). The fitted value of B^* is 28 T and 22 T for $x = 0.43$ and 0.48 samples respectively, which is larger than that of MgB_2 , ~ 10 T [16]. The higher B^* value indicates less suppression of the superconductivity by magnetic fields, which is consistent with the high H_{C2} in $\text{FeSe}_{1-x}\text{Te}_x$ superconductors. The irreversibility fields of $\text{FeSe}_{1-x}\text{Te}_x$ were investigated by means of temperature-dependent magnetization measurements [18]. A power-law temperature dependence, typically for cuprate superconductors, was observed and a high irreversibility field is expected at temperature well below T_C [18].

Figure 4. The activation energy of the vortex, U_0 , as a function of applied magnetic field for $x = 0.43$ (solid circles) and $x = 0.48$ (open circles) $\text{FeSe}_{1-x}\text{Te}_x$ films, in this work. The magnetic field dependence of U_0 follows a formula $U_0 = \alpha(1 - B/B^*)^2$ (solid lines). The fitted value of B^* is 28 T and 22 T for $x = 0.43$ and 0.48 samples respectively. The data for $\text{NdFeAsO}_{0.7}\text{F}_{0.3}$ (up-pointing triangles) [9] and $\text{Fe}_{1.03}\text{Te}_{0.55}\text{Se}_{0.45}$ (down-pointing triangles) [10] are also included for comparison.

Figure 5(a) shows the log-log plot of the E - J curves at different temperatures for the $x = 0.56$ film under a 7 T magnetic field. The data temperature ranges from 9.15 to 5.55 K, corresponding to a $T/T_C^{\text{offset}}(B = 0 \text{ T})$ range from 0.8 to 0.5. The E - J curve changes from having a positive curvature to having a negative curvature with decreasing temperature. This phenomenon can be well described by the vortex glass (VG) theory [19]. According to the VG theory, at the vortex glass transition temperature T_g , the E - J curve follows a power-law relation. Above and below T_g the curvature of E - J curve in the log-log plot changes from positive to negative. The T_g in figure 5(a) can be identified as 7.55 K, shown as a dashed line. In addition, the E - J curves can be scaled by using the formulas

$$\frac{E}{J(T - T_g)^{\nu(z+2-D)}} = f_{\pm} \left(\frac{J}{|T - T_g|^{\nu(D-1)}} \right), \quad (2)$$

where f_+ and f_- represent the function above and below T_g , respectively, and D is the dimensionality. According to the three-dimensional VG theory [19], the scaling parameter ν and z was suggested to be 4–7 and 1–2 respectively. The experiments on YBCO films reported the values of $\nu = 1.7$ and $z = 4.8$ [19]. The quasi-2D vortex glass systems were demonstrated in $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_x$ films and MgB_2 films by choosing $D = 2$ to get reasonable scaling parameters [20, 21]. The upper inset of figure 5(a) presents the scaled versions of the E - J curves in figure 5(a) at temperature near T_g . A good

scaling behavior of the E - J curves with critical exponents of $\nu = 1.2$ and $z = 4.0$ is obtained with $D = 3$, suggesting a three-dimensional system. Furthermore, the E - J relation obeys a power law at T_g with an exponent of $(z + 1)/(D - 1)$. The slope of the E - J curve at T_g in figure 5(a) is 2.45, which is almost the same as the value of 2.5 calculated using the parameters from scaling results. As $T \rightarrow T_g$, the linear resistivity vanishes with the form $\rho_{\text{lin}} \approx (T - T_g)^{\nu(z+2-D)}$, as shown in the lower inset. The linear fitting slope in the lower inset is 3.58, which is also in good agreement with the calculated value of 3.6. The VG transition temperature decreases with increasing magnetic field, from $T/T_C^{\text{offset}}(B = 0 \text{ T}) = 0.88$ under $B = 0 \text{ T}$ to $T/T_C^{\text{offset}}(B = 0 \text{ T}) = 0.7$ under $B = 7 \text{ T}$. The critical exponents ν and z are insensitive to the magnetic field strength within ranges of 1.35 ± 0.15 and 4.25 ± 0.25 .

For the $x = 0.85$ sample, the VG transition can also be observed under low magnetic field. Figure 5(b) presents the result under nominally zero magnetic field for the system. The scaled E - J curves and the resistivity vanishing as a power law at T_g are shown in the upper and lower insets of figure 5(b). However, the E - J curve at temperature down to 4 K still shows Ohmic behavior under high magnetic fields, which is consistent with the resistivity measurement results in figure 1(e).

Our results strongly suggest more effective pinning centers in samples with higher Te substitution level. According to the x-ray diffraction and SEM observations the crystallinities of these films, except for the $x = 0.85$ film, are similar. Therefore, we do not expect the grain boundaries to play important roles in the observed vortex pinning. It was pointed out from a neutron experiment [22] that there exists extra Fe located in interstitial sites. X-ray refinement results also demonstrated more excess Fe and greater suppression of the superconductivity in higher Te substitution samples [23–25]. Hence, the observation of a stronger pinning effect in the high Te substitution samples most likely originates from the excess Fe.

4. Conclusion

In conclusion, the vortex states of $\text{FeSe}_{1-x}\text{Te}_x$ superconducting thin films have been investigated. For higher Te substitution, $x = 0.56$ and 0.85 in this paper, a vortex glass transition was observed and the E - J curves can be well scaled on the basis of 3D vortex glass theory. The critical exponents ν and z are insensitive to the external magnetic field. The E - J characteristics of low Te substitution $\text{FeSe}_{1-x}\text{Te}_x$ films, $x < 0.48$ in this paper, still follow the thermally activated flux flow model even at $T/T_C^{\text{offset}}(B = 0 \text{ T}) = 0.4$ –0.5. The activation energy is found to be in the order of $100k_B T$ and the irreversibility field B_{irr} (or a Kramer scaling field B_K) is found to be a factor of 2 larger than those of the MgB_2 samples. Our results reveal enhancement of the flux pinning as Te is substituted, which is consistent with the tendency of excess Fe concentration. Our results suggest that $\text{FeSe}_{1-x}\text{Te}_x$ samples have great potential for practical applications.

Figure 5. (a) The E – J curves of $\text{FeSe}_{1-x}\text{Te}_x$, $x = 0.56$, thin film under a magnetic field of 7 T. The temperature ranges from 9.15 K to 5.55 K with a step of 0.2 K. The dashed line, a guide to the eye, indicates the vortex glass transition temperature $T_g = 7.55$ K, corresponding to the reduced temperature $T/T_C^{\text{offset}} = 0.7$. The upper inset displays the corresponding scaled E – J curves with $\nu = 1.2$, $z = 4.0$, and the dimension $D = 3$. The ρ_{lin} vanishes as a power law as T approaches T_g , as shown in the lower inset. (b) The E – J curves of $\text{FeSe}_{1-x}\text{Te}_x$, $x = 0.85$, thin film at zero magnetic fields. The temperature ranges from 8.9 to 4.2 K. The dashed line indicates the vortex glass transition temperature $T_g = 7.35$ K, corresponding to $T/T_C^{\text{offset}} = 0.7$. The upper and lower insets show the corresponding scaled E – J curves and the logarithmic plot of ρ_{lin} versus $(T - T_g)$, respectively.

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References

- [1] Kamihara Y, Watanabe T, Hirano M and Hosono H 2008 *J. Am. Chem. Soc.* **130** 3296
- [2] Chen X H, Wu T, Wu G, Liu R H, Chen H and Fang D F 2008 *Nature* **453** 761
- [3] Hsu F C *et al* 2008 *Proc. Natl Acad. Sci. USA* **105** 14262
- [4] Yuan H Q, Singleton J, Balakirev F F, Baily S A, Chen G F, Luo J L and Wang N L 2009 *Nature* **457** 565
- [5] Lee S *et al* 2010 *Nature Mater.* **9** 397
- [6] Zhang Y *et al* 2011 *Appl. Phys. Lett.* **98** 042509
- [7] Katase T, Ishimaru Y, Tsukamoto A, Hiramatsu H, Kamiya T, Tanabe K and Hosono H 2010 *Supercond. Sci. Technol.* **23** 082001
- [8] Kim H J, Liu Y, Oh Y S, Khim S, Kim I, Stewart G R and Kim K H 2009 *Phys. Rev. B* **79** 014514
- [9] Jaroszynski J *et al* 2008 *Phys. Rev. B* **78** 174523
- [10] Ge J, Cao S, Shen S, Yuan S, Kang B and Zhang J 2010 *Solid State Commun.* **150** 1641
- [11] Liu Y, Kremer R K and Lin C T 2010 *Europhys. Lett.* **92** 57004
- [12] Wu M K *et al* 2009 *Physica C* **469** 340
- [13] Wang M J *et al* 2009 *Phys. Rev. Lett.* **103** 117002
- [14] Anderson P W 1962 *Phys. Rev. Lett.* **9** 309
- [15] Anderson P W and Kim Y B 1964 *Rev. Mod. Phys.* **36** 39
- [16] Ketterton J B and Song S N 1999 *Superconductivity* (New York: Cambridge university press)
- [17] Thompson J R, Sorge K D, Cantoni C, Kerchner H R, Christen D K and Paranthaman M 2005 *Supercond. Sci. Technol.* **18** 970
- [18] Kaushik S D, Braccini V and Patnaik S 2008 *Pramana J. Phys.* **71** 1335
- [19] Bende M *et al* 2010 *Phys. Rev. B* **81** 224520
- [20] Fisher M P A 1989 *Phys. Rev. Lett.* **63** 1511
- [21] Fisher D S, Fisher M P A and Huse D A 1991 *Phys. Rev. B* **43** 130
- [22] Yamasaki H, Endo K, Kosaka S, Umeda M, Yoshida S and Kajimura K 1994 *Phys. Rev. B* **50** 12959
- [23] Yang H, Jia Y, Shan L, Zhang Y, Wen H H, Zhuang C, Liu Z, Li Q, Cui Y and Xi X 2007 *Phys. Rev. B* **76** 134513
- [24] Bao W *et al* 2009 *Phys. Rev. Lett.* **102** 247001
- [25] Sales B C, Sefat A S, McGuire M A, Jin R Y and Mandrus D 2009 *Phys. Rev. B* **79** 094521
- [26] Katayama N *et al* 2010 *J. Phys. Soc. Japan* **79** 113702
- [27] Bende M, Babkevich P, Katrych S, Gvasaliya S N, Pomjakushina E, Conder K, Roessli B, Boothroyd A T, Khasanov R and Keller H 2010 *Phys. Rev. B* **82** 212504