Magneto-optical characterizations of FeTe$_{0.5}$Se$_{0.5}$ thin films with critical current density over 1 MA cm$^{-2}$

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Received 30 May 2014, revised 16 October 2014
Accepted for publication 23 October 2014
Published 3 December 2014

Abstract

We performed magneto-optical (MO) measurements on FeTe$_{0.5}$Se$_{0.5}$ thin films grown on LaAlO$_3$ (LAO) and Yttria-stabilized zirconia (YSZ) single-crystalline substrates. These thin films show superconducting transition temperature $T_c$ $\sim$ 19 K, 4 K higher than the bulk sample. Typical roof-top patterns can be observed in the MO images of thin films grown on LAO and YSZ, from which a large and homogeneous critical current density $J_c$ over $1 \times 10^6$ A cm$^{-2}$ at 5 K was obtained. Magnetic flux penetration measurement reveals that the current is almost isotropically distributed in the two thin films. Compared with bulk crystals, FeTe$_{0.5}$Se$_{0.5}$ thin film demonstrates not only higher $T_c$, but also much larger $J_c$, which is attractive for applications.

Keywords: iron-based superconductors, thin film, Magneto-optical imaging

(Some figures may appear in colour only in the online journal)

1. Introduction

After the discovery of superconductivity in iron-based superconductors (IBSs) in 2008 [1], extensive research has revealed that this class of superconductors has fascinating fundamental properties for applications [2]. The upper critical field $H_{c2}$ reaches over 50 T in all field directions, and the anisotropy is moderate, much smaller than the cuprate superconductors. Accompanying small anisotropy, the superconducting transition under magnetic fields does not show an appreciable broadening, and the separation between $H_{c2}$ and the irreversibility field is much narrower than cuprates [3]. Among the family of IBSs, FeTe$_{1-x}$Se$_x$ has some practical advantages. Although the $T_c$ is typically below 20 K, it exhibits lower anisotropy $\sim$ 2 with $H_{c2} \sim$ 50 T [4, 5]. On the other hand, its simple structure and less toxic nature are also preferable for fabricating wires and thin films, although the critical current density $J_c$ in FeTe$_{0.5}$Se$_{0.5}$ bulks and wires [6, 7] is still much lower than that of Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ wires [8]. Our previous reports have demonstrated that high-quality FeTe$_{0.5}$Se$_{0.5}$ thin films can maintain a large $J_c$ over $10^6$ A cm$^{-2}$ at 4.2 K. Even under the field of 30 T, $J_c$ s are still exceeding $10^5$A cm$^{-2}$ [9, 10].

For the practical application of superconducting wires and tapes, local characterizations of $J_c$ distribution are very important. Magneto-optical (MO) imaging has proven extremely powerful for such purposes. It can visualize the spatial distribution of $J_c$, and is widely used to study the connectivity between grains in IBSs [6, 11–16]. In this report, we carefully studied the critical current density and its distribution in FeTe$_{0.5}$Se$_{0.5}$ thin films by MO imaging for the first time.
zirconia (YSZ) show large, homogeneous, and almost isotropic $J_c$ over $1 \times 10^6$ A cm$^{-2}$ at 5 K.

2. Experimental details

FeTe$_{0.5}$Se$_{0.5}$ thin films with thickness about 1000 Å were grown using pulsed laser deposition. Films were deposited on CeO$_2$ buffered single-crystalline substrates LAO and YSZ. Details of the growth conditions have been reported in our previous publications [17, 18]. X-ray diffraction patterns reported in our previous paper show that only (00$\bar{l}$) peaks from the FeTe$_{0.5}$Se$_{0.5}$ thin films are present [10]. The inset of figure 1 shows a typical $\phi$ scan of the (101) peak from the film grown on YSZ, manifesting a sharp full width at half maximum (FWHM, $\Delta \phi$) of 1.3$^\circ$. The preceding results indicate the good in-plane alignments of FeTe$_{0.5}$Se$_{0.5}$ thin films without grain boundaries.

Resistivities were measured using the standard four-probe method. Magnetization measurements were performed using a commercial superconducting quantum interference device (SQUID). MO images were obtained by using the local field-dependent Faraday effect. For MO measurements, the films were cut into rectangular shapes with dimensions about $1 \times 1 \text{ mm}^2$ for those grown on LAO, and $0.85 \times 0.78 \text{ mm}^2$ for those grown on YSZ. An in-plane magnetized garnet indicator film was placed in direct contact with the surface of the sample and the image of the reflected light, which is related to the local magnetic induction around the sample, was captured by using an optical microscope and a cooled-CCD camera (ORCA-ER, Hamamatsu). The sample was cooled by a He-flow-type cryostat (Microstat-HR, Oxford Instruments). A sketch of the MO imaging system can be found in our previous report [19]. To enhance the visibility and remove unnecessary contrasts coming from the defects of the garnet film, differential MO images were obtained by taking the difference between the two integrated images at $H = H_t$ and $H = 0$ [11, 20].

3. Results and discussion

Figure 1 shows the resistive transitions of FeTe$_{0.5}$Se$_{0.5}$ thin films grown on LAO and YSZ substrates together with that of a single crystal. $T_c$ of the single crystal is $\sim$15 K, similar to the previous reports [15, 21]. The thin films grown on LAO and YSZ single-crystalline substrates both show $T_c$ $\sim$19 K, 4 K higher than the bulk sample. The behavior of higher $T_c$ observed in thin films than that of bulk samples is common in iron chalcogenides. Strain in the films from substrates is thought to be a contributing factor for higher $T_c$ observed in the superconducting iron chalcogenide films. [17, 18, 22].

MO images in the remanent state are prepared by a applying magnetic field, large enough to totally penetrate the sample, along $c$-axis, and removing it after zero-field cooling. Typical MO images of FeTe$_{0.5}$Se$_{0.5}$ thin film grown on LAO substrate from 5 to 16 K are shown in figures 2(a)–(e). The MO image manifests a typical roof-top pattern, similar to that observed in high-quality FeTe$_{0.5}$Se$_{0.4}$ single crystal [15, 23–26], indicating a nearly uniform current flow in the thin film. To directly observe the distribution of $J_c$ in the film, we convert the MO images into the current distribution with a thin-sheet approximation by using the fast Fourier transform process to simplify Biot–Svart law [27, 28]. A typical image of the distribution of the modulus of $J_c$ at 5 K is shown in figure 2(f), which again manifests that the current is homogeneously distributed in most parts of the thin film. Figure 2(g) shows the profiles of the magnetic induction along the dashed line in figure 2(a) at different temperatures. From this profile, the critical current density $J_c$ for the thin film can be roughly estimated from the following formula:

$$\Delta B = \frac{8J_c \cdot 2d}{c} \int_0^a \frac{\alpha^2}{\sqrt{z^2 + \frac{2\alpha^2}{(2z^2 + \alpha^2)}}} \, dz,$$

where $\Delta B$ is the trapped field in the film, $2d$ and $2a$ are the thickness and length/width of the sample, respectively, $z_g$ is the distance from the film surface to the MO indicator, and $c$ is the velocity of light, which is equal to 10 in the practical unit (derivation of the formula can be seen in the supplementary material). Substituting $z_g = 3 \mu$m from our experience in MO imaging [29], and $a = 500 \mu$m into the preceding formula, the integral part can be numerically calculated as 3.471. Thus, the magnetic field along the $z$-axis can be simply estimated as: $J_c = \Delta B/(2d \times 2.78)$. Temperature dependence of $J_c$ obtained from the MO image is plotted in figure 2(h). Here, $J_c$ at 5 K shows a large value of $\sim 1.1 \times 10^6$ A cm$^{-2}$, which is close to that obtained from MHLs by the extended Bean model [30] (shown as the open
circle in figure 2(h)).

\[ J_c = \frac{\Delta M}{2a(1 - a/b)} \]  

where \( \Delta M \) is \( M_{\text{down}} - M_{\text{up}} \), \( M_{\text{up}} \) [emu cm\(^{-3}\)] and \( M_{\text{down}} \) [emu cm\(^{-3}\)] is the magnetization when sweeping fields up and down, respectively, and \( 2a \) [cm] and \( 2b \) [cm] are sample widths (\( a < b \)).

Figure 3 (a)–(e) show the MO images in the remanent state of a FeTe\(_{0.5}\)Se\(_{0.5}\) thin film grown on the LAO substrate at (a) 5, (b) 8, (c) 10, (d) 14, and (e) 16 K. (f) Temperature dependence of \( J_c \) derived from MO, together with that obtained from magnetic hysteresis loops (MHLs) (shown as the open circle).

\[ \Delta M = - J M_{\text{aa}} b^2 (1/3) \]

\( M_{\text{up}} \) [emu cm\(^{-3}\)] and \( M_{\text{down}} \) [emu cm\(^{-3}\)] is the magnetization when sweeping fields up and down, respectively, and \( 2a \) [cm] and 2b [cm] are sample widths (\( a < b \)).

Figure 3 (a)–(e) show the MO images in the remanent state of a FeTe\(_{0.5}\)Se\(_{0.5}\) thin film grown on the LAO substrate at temperatures ranging from 5 to 16 K. Similar to the film grown on the LAO substrate, MO images at temperatures lower than 14 K show typical roof-top patterns, indicating the homogeneous distribution of critical current density in the thin film grown on the YSZ substrate. The homogeneous distribution of \( J_c \) can be directly observed in its spatial distribution, as shown in figure 3(f). The superconductivity observed above 15 K seems less homogeneous because only parts of the thin film can trap field, as shown in figure 3(e). Profiles of the magnetic induction along the dashed line in
figure 3(a) at different temperatures are shown in figure 3(g). Here, \(J_c\)s at different temperatures are roughly estimated and shown in figure 3(h). The value of \(J_c\) reaches \(\sim 1.4 \times 10^6\) A cm\(^{-2}\) at 5 K, and is also close to the global \(J_c\) obtained from MHLs shown as the open square in figure 3(h).

Figures 4(a)–(c) and (d)–(f) reveal the penetration of magnetic flux at 5 K for FeTe\(_{0.5}\)Se\(_{0.5}\) thin films grown on LAO and YSZ, respectively. Obviously, flux gradually penetrates the two thin films with increasing applied field. More importantly, the typical current discontinuity lines (d-line), which cannot be crossed by vortices, can be directly observed and marked by the dashed line in figures 4(c) and (f). By measuring the angles of the discontinuity line for the rectangular film, the in-plane anisotropy of the current
densities can be easily estimated [31]. In this case, the angle $\theta$ is close to 45°, indicating that the critical current density in the $ab$-plan is almost isotropic. Schematics of the current distributions are presented by the dotted lines.

Here, we should point out that we have performed extensive structural and chemical analyses of these films at room temperature by both synchrotron-based x-ray diffraction and analytical transmission electron microscope equipped with nano-probe electron energy loss spectroscopy. We found no evidence of structural and chemical inhomogeneities of these films at the macroscopic level. However, we did notice some minor inhomogeneities in the trapped magnetic flux patterns from MO images. Although the observed inhomogeneities are too small to have a meaningful impact on our determination of the local critical current density of these films, they do suggest that these films are very sensitive to small perturbations, which are presumably related to the strain built in the films. There are several possible ways to induce strain in thin films. Among them are epitaxial strain due to slightly lattice constant mismatch between the film and substrates, strain induced during the sample cool-down from processing temperature (400°C) to room temperature or from room temperature to low temperature for MO experiments. The latter type of thermally induced strain tends to be less macroscopically uniform, and hence may cause variations of $T_c$ and $J_c$ across the entire film that might affect the uniformity of the flux patterns.

In figure 5, we compare the temperature dependence of $J_c$ for FeTe$_{0.5}$Se$_{0.5}$ thin films grown on LAO and YSZ substrates together with that of a high-quality FeTe$_{0.6}$Se$_{0.4}$ single crystal [15]. The values of $J_c$ in FeTe$_{0.5}$Se$_{0.5}$ thin films are close to those of the Nb$_3$Sn wires. Considering the much lower $H_{c2}$ of Nb$_3$Sn, FeTe$_{0.5}$Se$_{0.5}$ may be a respectable candidate for applications at the liquid helium temperatures. In addition, we previously reported the successful fabrication of coated FeTe$_{0.5}$Se$_{0.5}$ tapes with $J_c$ over $10^6$ A cm$^{-2}$ under zero field, and over $1 \times 10^5$ A cm$^{-2}$ under 30 T magnetic field at 4.2 K [10]. All these results show that FeTe$_{0.5}$Se$_{0.5}$ is suitable for high-field applications.

4. Conclusions

In conclusion, MO images were taken on FeTe$_{0.5}$Se$_{0.5}$ thin films grown on LAO and YSZ substrates together with that of a FeTe$_{0.6}$Se$_{0.4}$ single crystal [15]. The values of $J_c$ in FeTe$_{0.5}$Se$_{0.5}$ thin films are close to those of the Nb$_3$Sn wires. Considering the much lower $H_{c2}$ of Nb$_3$Sn, FeTe$_{0.5}$Se$_{0.5}$ may be a respectable candidate for applications at the liquid helium temperatures. In addition, we previously reported the successful fabrication of coated FeTe$_{0.5}$Se$_{0.5}$ tapes with $J_c$ over $10^6$ A cm$^{-2}$ under zero field, and over $1 \times 10^5$ A cm$^{-2}$ under 30 T magnetic field at 4.2 K [10]. All these results show that FeTe$_{0.5}$Se$_{0.5}$ is suitable for high-field applications.

Figure 4. MO images of flux penetrations into the FeTe$_{0.5}$Se$_{0.5}$ thin films grown on LAO and YSZ substrates at 5 K under different magnetic fields. The red dashed lines in (c) and (f) show the current discontinuity lines, which cannot be crossed by vortices. The yellow dotted lines are the schematics of the current distributions in the thin films.

Figure 5. Temperature dependence of $J_c$ obtained from MO and MHLs for FeTe$_{0.5}$Se$_{0.5}$ thin films grown on LAO and YSZ substrates, together with that of a FeTe$_{0.6}$Se$_{0.4}$ single crystal [15].
and field-independent $J_c$, very high upper critical field, and relatively low cost make FeTe$_{0.5}$Se$_{0.5}$ an attractive candidate for applications at liquid helium temperatures, especially under a high magnetic field.

Acknowledgements

Yue Sun gratefully appreciates the support from the Japan Society for the Promotion of Science. The work at the University of Tokyo was supported by the Japan-China Bilateral Joint Research Project of the Japan Society for the Promotion of Science. The work at Brookhaven National Laboratory was supported by the U.S. Department of Energy, Office of Basic Energy Science, Materials Sciences and Engineering Division, under contract no. DEAC0298CH10886.

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