Upper critical field, lower critical field and critical current density of FeTe$_{0.60}$Se$_{0.40}$ single crystals

C S Yadav$^1$ and P L Paulose$^1$
Department of Condensed Matter Physics and Material Sciences, Tata Institute of Fundamental Research, Colaba, Mumbai-400005, India
E-mail: csyadav@tifr.res.in and paulose@tifr.res.in

New Journal of Physics 11 (2009) 103046 (10pp)
Received 21 June 2009
Published 26 October 2009
Online at http://www.njp.org/
doi:10.1088/1367-2630/11/10/103046

Abstract. Transport and magnetic studies are performed on high-quality FeTe$_{0.60}$Se$_{0.40}$ single crystals to determine the upper critical field ($H_{c2}$), lower critical field ($H_{c1}$) and the critical current density ($J_c$). The value of the upper critical field $H_{c2}$ is very large, whereas the activation energy determined from the slope of the Arrhenius plots is found to be lower than that in the FeAs122 superconductors. The lower critical field is determined in the ‘ab’ and ‘c’ directions of the crystals, and found to have anisotropy $\Gamma(=H_{c1}/H_{c1\parallel ab}) \sim 4$. The magnetic isotherms measured up to 12 T show the presence of fishtail behavior. The critical current density at 1.8 K of the single crystals is found to be almost the same in both the ‘ab’ and ‘c’ directions in the low-field regime.

Contents

1. Introduction 2
2. Experimental methods 2
3. Results and discussions 2
4. Conclusions 9
Acknowledgment 9
References 9

$^1$ Authors to whom any correspondence should be addressed.
1. Introduction

The discovery of superconductivity in the Fe-based oxypnictide compounds has enriched and opened up new horizons in the field of superconductivity [1]. The tetragonal compounds FeSe and FeTe$_{1-x}$Se$_x$ have a simpler structure than FeAs-based superconductors, where the Fe(Te/Se) layers stack along the c-axis, and have a transition temperature ($T_c$) as high as 15 K [2]–[9]. Pressure studies on the FeSe compounds show an increase in the $T_c$ up to 36 K at 38 GPa pressure [2, 10, 11]. Although the $T_c$ in these compounds is much less than that of FeAs-based superconductors, the simplicity of structure and similarity in the Fermi surface make them a potentially useful material to help understand the superconducting mechanism in the Fe-based oxypnictides. The Fermi surface of FeS, FeSe and FeTe is very similar to that of FeAs-based superconductors, with the cylindrical hole and electron sections at the center and the corner of the Brillouin zone, respectively [12]. The end member FeTe in the FeTe$_{1-x}$Se$_x$ series is antiferromagnetic below 65 K and shows a simultaneous structural transition [4, 5, 8, 9]. Among the Fe mono-chalcogenide compounds, only FeSe shows superconductivity ($T_c$ = 8 K), but it is difficult to prepare FeSe in pure form as 1–2% impurity of the Fe$_7$Se$_8$ hexagonal phase forms along with the superconducting tetragonal FeSe phase [2, 4], [6]–[8]. The substitution of Te at the Se site in FeSe increases the $T_c$, showing a maximum Se concentration of close to 40% [4, 5, 9].

In this paper, we have estimated the upper critical field ($H_{c2}$), activation energy ($U_0$), lower critical field ($H_{c1}$) and the critical current density ($J_c$) of a high-quality single crystal of FeTe$_{0.60}$Se$_{0.40}$ with a superconducting volume fraction of more than 95%. We have also observed the fishtail behavior in the high-field magnetization loop at temperatures below $T_c$ for both directions.

2. Experimental methods

The single crystals of FeTe$_{0.60}$Se$_{0.40}$ compound were prepared by the chemical reaction of the elements (Fe chunk of 99.999% purity, Te powder of 99.99% purity and Se powder of 99.98% purity) in the stochiometric proportion, inside a sealed quartz tube under vacuum. The charge was slowly heated to 950°C at the rate of 50 °C h$^{-1}$ and kept for 12 h before cooling down to 400 °C at the rate of 6 °C h$^{-1}$, and then the furnace was cooled to room temperature to grow the crystals.

The Magnetotransport measurements were performed using a Quantum Design PPMS (physical properties measurement system). The specific heat of crystals was measured using the relaxation technique in the PPMS. Ac susceptibility measurements, and low-field dc magnetization were carried out using a superconducting quantum interference device (SQUID) magnetometer and high magnetic field measurements were performed using a 12 T vibrating sample magnetometer (Oxford Instruments).

3. Results and discussions

The crystals were found to be very shiny, grown along the ‘ab’ plane, and were very easy to cleave along this plane. The x-ray diffraction (XRD) analysis performed on the powdered sample confirmed the compound to be in a single tetragonal phase (space group P4/nmm), with the lattice parameters ‘$a$’ = 3.798 Å and ‘$c$’ = 6.058 Å. The compositional analysis by
energy dispersive absorption x-ray spectroscopy (EDAX), showed the crystals to be formed in the stochiometric ratio. The XRD pattern of crystal flakes shows peaks only at the angles corresponding to the \( \{001\} \) planes, confirming the orientation of the flakes along the \( ab \) plane. To further check the quality of the crystal, the transmission electron microscope (TEM) diffraction pattern (shown in figure 1) was recorded, which again confirmed the tetragonal phase and growth of the crystal along the \( ab \) plane.

In figure 2, we have shown the resistivity data of FeTe\(_{0.60}\)Se\(_{0.40}\) single crystals for a magnetic field parallel to ‘\( c \)’ and the electrical current in the ‘\( ab \)’ plane. The room temperature resistivity is about 0.9 m\( \Omega \) cm. It is metallic below about 150 K and superconducts with a \( T_c \) onset of 15.3 K (inset of figure 2). At zero magnetic field, the transition width is 0.5 K, which is considerably broadened to 2.3 K at 14 T field. However the \( T_c \) onset is not affected very much by the magnetic field as reported in the case of cuprate superconductors. Like the two-dimensional cuprate superconductors, the FeAs-based layered systems are also reported to have a very high critical field [13]. In figure 3, we have plotted the \( H-T \) phase diagram for the crystals, corresponding to the temperatures where the resistivity drops to 90% of the normal state resistivity \( \rho_n \), (where \( \rho_n \) is taken at temperature \( T = 16 \) K), 50% of \( \rho_n \) and 10% of \( \rho_n \). Since the transition temperature does not shift much towards the low temperatures, it indicates a very high value of \( H_{c2}(0) \) at the zero temperature. The linear extrapolation of the field axis lines at \( T = 0 \) K gave the values of high critical field \( H_{c2}(0) \) as 184, 88 and 69 T, corresponding to the transition temperatures taken at the point of 90, 50 and 10% of \( \rho_n \), respectively. Applying the Werthamer–Helfand–Hohenberg (WHH) formula

\[
\mu_0 H_{c2}(0) = -0.693 \mu_0 \left( \frac{dH_{c2}}{dT} \right)_{T_c} T_c
\]
to the $H$–$T$ phase diagram shown in figure 3, the $H_{c2}(0)$ were found to be 126, 65 and 51 T corresponding to the points 90% of $\rho_n$, 50 and 10% of $\rho_n$, respectively. Using these zero temperature value of $H_{c2}$, the corresponding value of $\mu_0 H_{c2}/k_B T_c$ comes out to be 8.21, 4.26 and 3.30 T K$^{-1}$, which are much higher than the Pauli limit for $\mu_0 H_{c2}/k_B T_c = 1.84$ T K$^{-1}$ in the case of singlet pairing and weak spin–orbit coupling [13, 14]. This indicates the unconventional nature of the superconductivity. In order to approximate the superconducting parameters, we have used the Ginzburg–Landau (GL) formula for the coherence length ($\xi$),

$$\xi = (\Phi_0/2\pi \mu_0 H_{c2})^{1/2},$$

where $\Phi_0 = 2.07 \times 10^{-7}$ Oe cm$^2$, the coherence length $\xi$ at the zero temperature was calculated as 16.2, 22 and 25.5 Å for the $H_{c2}$ at 90, 50 and 10% of $\rho_n$, respectively.

The Arrhenius plot for the FeTe$_{0.60}$Se$_{0.40}$ in figure 4 shows that the electrical resistivity is thermally activated in the region of resistivity between $2 \times 10^{-4}$ and $2 \times 10^{-6}$ $\Omega$ cm. The activation energy $U_0$ is determined from the slope of the curve in this linear region using the formula $\rho(T, H) = \rho_0 \exp(-U_0/k_B T)$. The magnetic field versus the activation energy $U_0$ plot shown in the inset of figure 4 suggests the different power-law dependence on magnetic field $U_0 \propto H^{-\alpha}$, with $\alpha = 0.10$ for $0 < H < 6$ T and $\alpha = 0.57$ for $6 T < H < 14$ T. Similar power-law dependence has also been observed for other superconducting compounds, e.g. Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$, MgB$_2$, SmFeAsO$_{0.9}$F$_{0.1}$ and NdFeAsO$_{0.82}$F$_{0.18}$ [15]–[18]. The activation energy varies linearly from 710 to 1490 K for the magnetic field of $H = 14$ and 0 T, respectively.

The temperature dependence for the specific heat and the ac magnetic susceptibility of FeTe$_{0.60}$Se$_{0.40}$ are shown in figure 5(a). The inset of this figure contains the $M$–$H$ hysteresis

**Figure 2.** Temperature dependence of the resistivity of FeTe$_{0.60}$Se$_{0.40}$ single crystals, measured in the magnetic fields (from right to left) $H = 0$, 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13 and 14 T. The semi-metallic behavior of resistivity in the normal region is shown in the inset of the figure.
Figure 3. The upper critical field versus temperature phase diagram is shown for the points where electrical resistivity drops to 90, 50 and 10% of $\rho_n$, shown by $T_{\text{onset}}$ and $T_{\text{mid}}$ and $T_{\text{offset}}$. $\rho_n$ is the value of resistivity taken in the normal state at 16 K.

Figure 4. The Arrhenius plot of the resistivity for FeTe$_{0.60}$Se$_{0.40}$ single crystals for (left to right) $H = 0, 1, 2, 3, 4, 5, 6, 8, 9, 10, 11, 12, 13$ and 14 T. The inset of the figure shows variations of $U_0$ with the magnetic field.
Figure 5. (a) The temperature dependence of the specific heat and the ac susceptibility of FeTe$_{0.60}$Se$_{0.40}$ crystals. The inset shows the $M$–$H$ loop at 1.8 K. (b) The field dependence of the initial magnetization isotherms is plotted for different temperatures. The dotted line gives the linear fit to the low-field $MH$ curve at 1.8 K. (c) Deviation of $M$, from the linear low-field $M$–$H$ slope ($\Delta M$) is plotted for different temperatures. Inset of figure (c) shows the point of deviation of $\Delta M$ from the zero base line for $T = 1.8$ K, giving the value of the first-penetration field. (d) The lower critical field $H_{c1}$ measured for $H \parallel c$ and $H \parallel ab$ shows a positive curvature all along the temperature region. The anisotropy parameter $\Gamma (= (H_{c1} \parallel c)/(H_{c1} \parallel ab))$ was estimated to be $\sim 4$ at 1.8 K.

loop measured at 1.8 K. Although the ac susceptibility data show almost complete expulsion of the magnetic flux in the Meissner state ($\sim 95\%$ of the superconducting volume), the kink in the specific heat is not very sharp, as expected in the case of second-order superconducting transition [19]. The Meissner value of the diamagnetic susceptibility is almost constant below 10 K. The value of ac susceptibility in the region 1.8–10 K is almost constant at $\chi = -1.35 \times 10^{-2}$ emu g$^{-1}$.

We measured the field dependence of the magnetization in the superconducting state at different temperatures, with the external magnetic field along the $ab$-basal plane and the $c$-axis. Figure 5(b) shows the different magnetization isotherms for the field direction along the $ab$ plane. The linear variation of the magnetization ($-M$), which is the signature of the Meissner state, is clearly seen in the low-field region. The lower critical field $H_{c1}$, as determined from the...
point of deviation from the linear $M$ versus $H$ of the dc magnetization data, was calculated to be
82 Oe for $H \parallel ab$ at $T = 1.8$ K. The low-field slope of the $M$–$H$ curve shown as a dotted line,
gives the value of susceptibility $\chi_{dc} = 1.15 \times 10^{-2}$ emu g$^{-1}$, which is very near to the value of
ac susceptibility $\chi_{ac} = 1.35 \times 10^{-2}$ emu g$^{-1}$ at 1.8 K (also shown in figure 5(a) in the Meissner
state.

For the accurate determination of the lower critical field we subtracted the value of
magnetization ($M_0$) obtained by the low-field magnetization slope from the magnetization ($M$)
for each isotherm [20, 21]. The deviation point of $\Delta M$ (i.e. $M$–$M_0$) versus field curve from the
zero base line gives the value of the first penetration field ($H_{c1}^*$), where the vortex starts entering
into the sample. The $\Delta M$ versus $H$ plots thus obtained for different temperatures are shown
in figure 5(c). The inset of the plot shows $\Delta M$ versus $H$ for $T = 1.8$ K. The lower critical field
$H_{c1}$ can be deduced from $H_{c1}^*$. For rectangle sample geometry, Brandt gave the relation between
$H_{c1}$ and $H_{c1}^*$ as $H_{c1}^* = H_{c1}/\tanh(0.36b/a)$, where ‘a’ and ‘b’ are the width and the thickness
of the samples, respectively [22]. Using this formula, we estimated the effective demagnetizing
factor $N_{eff} \sim 0.79$ for our sample with dimensions ‘a’ = 2.2 mm and ‘b’ = 0.25 mm.

As shown in figure 5(d), the $H_{c1}$ values for $H \parallel c$ and $H \parallel ab$ are highly temperature
dependent and show an upward trend with the negative curvature. A similar trend is reported
for FeAs-based superconductors such as Ba$_{0.66}$K$_{0.40}$Fe$_2$As$_2$ and SmFeAsO$_{0.9}$F$_{0.1}$. This has been
observed as not conforming to the single-band gap description of the mean-field theory, and
hence as evidence of two energy gaps, as with the MgB$_2$ superconductor [23]–[25]. The density-
functional study of FeSe and FeTe performed by Subedi et al showed that the band structure
of these compounds consists of a cylindrical electron Fermi surface at the zone corner, and two
concentric cylindrical hole surfaces at the zone center, indicating that the superconductivity in
this system results from two bands [12], and the upward curvature of $H_{c1}$ is dictated by both
the electrons and the heavy holes. The $H_{c1}$ values for our FeTe$_{0.60}$Se$_{0.40}$ crystals were found to
have an anisotropy ratio ($\Gamma = (H_{c1} \parallel c)/(H_{c1} \parallel ab)$) of 2–4 for the temperature range 1.8 K <
$T < 14$ K. This anisotropy is larger than that in PrFeAsO$_{1−x}$ and Sm$_{0.95}$La$_{0.05}$FeAs$_{0.85}$F$_{0.15}$
[26, 27].

Figures 6(a) and (b) show the $M$–$H$ loop in the positive field direction at several
temperatures in the magnetic field parallel to ab plane ($H \parallel ab$) and parallel to the c-axis ($H \parallel c$)
which was measured up to 12 T. The magnetization ‘$−M$’ goes through a first maximum on
increasing the magnetic field and shows a second peak before it finally collapses to zero near the
upper critical field $H_{c2}$. This second maximum is known as the fishtail effect in the literature
and has also been observed for crystals of LaSrCuO, YBCO, BSCCO, and more recently in
Ba(Fe$_{0.93}$Co$_{0.07}$)$_2$As$_2$ single crystals [28]–[31]. Although the origin of this behavior is not fully
explained yet, one model correlates it to the presence of some weakly superconducting or
non-superconducting regions that can act as the efficient pinning centers [29, 30]. It is also
propounded that the crossover from single to collective flux creep induces a slower magnetic
relaxation at the intermediate field and give rise to the second peak [29]–[31]. However, the
fishtail is strongly dependent on the sample orientation of the externally applied field, and for $H$
parallel to the ab-plane this feature is diminished.

Using the Bean model for the field-independent critical current density ($J_c$), it can be
calculated by the relation [32, 33]

$$ J_c = \frac{20 \Delta M}{a(1 − a/3b)}, $$

New Journal of Physics 11 (2009) 103046 (http://www.njp.org/)
Figure 6. The field dependence of magnetic isotherms measured up to the 12 T field for (a) the $H \parallel ab$ plane and (b) the $H \parallel c$-axis shows the fishtail-like feature at higher temperatures. The field dependence of the critical current density $J_c$ at different temperatures is plotted on the log–log scale for (c) the $H \parallel ab$-plane and (d) the $H \parallel c$-axis.

where $\Delta M = M_{\text{up}} - M_{\text{dn}}$, and $M_{\text{up}}$ and $M_{\text{dn}}$ are the magnetization while the magnetic fields are decreasing and increasing, respectively; $a$ and $b$ are the sample width ($a < b$). We took the sample dimensions as $a = 2.2$ mm, $b = 3$ mm and $a = 0.25$ mm, $b = 2.2$ mm for the $J_c$ calculation for $H \parallel ab$ and $H \parallel c$ respectively. It should be mentioned that although the Bean model is strictly applicable only in the case of field-independent critical current density, because the variation of $J_c$ is moderate up to 6 T for $H \parallel ab$ (up to 1 T for $H \parallel c$) it serves as a good approximation for the actual value.

The critical current density $J_c$ obtained for the FeTe$_{0.60}$Se$_{0.40}$ single-crystal sample for $H \parallel ab$ and $H \parallel c$ is shown in figures 6(c) and (d). The value of $J_c$ at low-field and at 1.8 K is almost identical at close to $1 \times 10^5$ A cm$^{-2}$ for both directions. The fishtail feature is also more clearly evident. Our value of $J_c$ agrees with the recent report by Taen et al for FeTe$_{0.61}$Se$_{0.39}$ crystals [33]. However, in an earlier report on Fe$_{1+y}$Te$_{1-x}$Se$_x$; $x = 0.133$, Hu et al reported an anisotropy in critical current density ($J_{c||ab}/J_{c||c}$) $\sim 5$ in their single crystal with a 10% superconducting volume fraction [34]. The current density $J_c$ values also compare well with those for the Co-doped BaFe$_2$As$_2$ superconductor [35].

New Journal of Physics 11 (2009) 103046 (http://www.njp.org/)
4. Conclusions

We have determined the upper critical field ($H_{c2}$), activation energy ($U_0$), lower critical fields ($H_{c1}$) and the critical current density ($J_c$) of FeTe$_{0.60}$Se$_{0.40}$ single crystals. The $H_{c2}$ values at $T = 0$ K, measured along the $ab$ plane, with extrapolation from the $H-T$ phase diagram, and using the WHH formula, are found to be very high. The activation energy shows linear dependence with the magnetic field. The $H-T$ phase diagram for $H_{c1}$ shows a positive curvature and does not saturate till 1.8 K. The lower critical field was found to be anisotropic, with the anisotropy parameter $\Gamma = (H_{c1\|c})/(H_{c1\|ab}) \sim 4$ at 1.8 K. The high-field $M-H$ behavior shows fishtail behavior, and is more pronounced in the $H \parallel c$ direction. The critical current density $J_c$ of the compound is found to be $1 \times 10^5$ A cm$^{-2}$ at low field and 1.8 K temperature, and appears to be isotropic in nature.

Acknowledgment

We acknowledge Manish Ghagh for his help with sample preparation and measurements.

References

[1] Kamihara Y, Hiramatsu H, Hirano M, Kawamura R, Yanagi H, Kamiya T and Hosono H 2006 J. Am. Chem. Soc. 128 10012
Kamihara Y, Hiramatsu H, Hirano M, Kawamura R, Yanagi H, Kamiya T and Hosono H 2008 J. Am. Chem. Soc. 130 3296
[2] Hsu F C et al 2008 Proc. Natl Acad. Sci. USA 105 14262
[3] Mizuguchi Y, Tomioka F, Tsuda S, Yamaguchi T and Takano Y 2008 Appl. Phys. Lett. 93 0152505
[4] Yeh K W et al 2008 Eur. Phys. Lett. 84 37002
[5] Fang M H, Pham H M, Qian B, Liu T J, Vehstedt E K, Liu Y, Spinu L and Mao Z Q 2008 Phys. Rev. B 78 224503
[6] Mcqueen T M et al 2009 Phys. Rev. B 79 014522
[7] Sales B C, Sefat A S, McGuire M A, Jin R Y and Mandrus D 2009 Phys. Rev. B 79 94521
[8] Chen G F, Chen Z G, Dong J, Hu W Z, Li G, Zhang X D, Zheng P, Luo J L and Wang N L 2009 Phys. Rev. B 79 140509
[9] Liu T J et al 2009 arXiv:0904.0824
[10] Medvedev S, McQueen T M, Trojan I, Palasyuk T, Eremets M I, Cava R J, Naghavi S, Casper F, Ksenofontov V, Wortmann G and Felser C 2009 arXiv:0903.2143
[11] Margadonna S, Takabayashi Y, Ohisi Y, Mizuguchi Y, Takano Y, Kagayama T, Nakagawa T, Takata M and Prassides K 2009 arXiv:0903.2204
[12] Subedi A, Zhang L, Singh D J and Du M H 2008 Phys. Rev. B 78 134514
[13] Wang Z S, Luo H Q, Ren C and Wen H H 2008 Phys. Rev. B 78 140501
[14] Clogston A M 1962 Phys. Rev. Lett. 9 266
[15] Palstra T T M, Batlogg B, Schneemeyer L F and Waszczak J V 1988 Phys. Rev. Lett. 61 1662
[16] Zhang Y Z, Ren Z A and Zhao Z X 2009 arXiv:0904.3625
[17] Zhang Y Z, Wang Z, Lu X F, Wen H H, de Marneffe J F, Deltour R, Jansen A G M and Wyder P 2005 Phys. Rev. B 71 052502
[18] Wang X L, Ghorbani S R, Dou S X, Shen X L, Yi W, Li Z C and Ren Z A 2008 arXiv:0806.1318
[19] Dong J K, Ding L, Wang H, Wang X F, Wu T, Wu G, Chen X H and Li S Y 2008 New J. Phys. 10 123031
[20] Balakrishnan G, Subramaniam C K, Paul D McK, Pinol S and Vijayaraghavan R 1991 Physica C 177 310
[21] Naito M, Matsuda A, Kitazawa K, Kambe S, Tanaka I and Kojima H 1990 Phys. Rev. B 41 4823

New Journal of Physics 11 (2009) 103046 (http://www.njp.org/)
[22] Brandt E H 1999 Phys. Rev. B 17 11939
[23] Ren C, Wang Z S, Luo H Q, Yang H, Shan L and Wen H H 2008 Phys. Rev. Lett. 101 257006
[24] Ren C, Wang Z S, Luo H Q, Yang H, Shan L and Wen H H 2009 Physica C 469 599
[25] Okazaki R et al 2009 Phys. Rev. B 79 064520
[26] Chang B C, Hsu C H, Hsu Y Y, Kei Z, Ruan K Q, Li X G and Ku H C 2008 Eur. Phys. Lett. 84 67014
[27] Lyard L, Szabo P, Klein T, Marcus J, Marcenat C, Kim K H, Kang B W, Lee H S and Lee S I 2004 Phys. Rev. Lett. 92 057001
[28] Bugoslavsky Yu V, Ivanov A L, Minakov A A and Vasyurin S I 1994 Physica C 233 67
[29] Elbaum L K, Civale L, Vinokur V M and Holtzberg F 1992 Phys. Rev. Lett. 69 2280
[30] Wei C D, Liu Z X, Ren H T and Xiao L 1996 Physica C 260 130
[31] Prozorov R et al 2008 Phys. Rev. B 78 224506
[32] Bean C P 1964 Rev. Mod. Phys. 36 31
[33] Taen T, Tsuchiya Y, Nakajima Y and Tamegai T 2009 arXiv:0906.1951
[34] Hu R, Warren J B and Petrovic C 2009 arXiv:0903.4430
[35] Nakajima Y, Taen T and Tamegai T 2009 J. Phys. Soc. Japan 78 023702