Weak localization in $\text{FeSe}_{1-x}\text{Te}_x$ superconducting thin films

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Received 18 October 2011, in final form 19 December 2011

Published 20 January 2012

Online at stacks.iop.org/SUST/25/035004

Abstract

We have investigated the magneto-resistivity (MR) of $\text{FeSe}_{1-x}\text{Te}_x$ superconducting films on MgO substrate. The MR of pure FeSe and slightly Te-substituted films demonstrates regular Lorentz-type magnetic field dependence, $\text{MR} \sim B^2$. In highly Te-substituted samples, however, negative MR contribution due to the weak-localization effect gradually dominates at low temperature, which is consistent with the evolution of the temperature dependence of resistivity from a metallic to a weakly semiconductor-like behavior. Furthermore, the negative MR weakens and turns positive as temperature approaches the superconducting transition temperature, which is evidence for the Maki–Thompson correction in the weak-localization regime. The experimental data can be described very well by the weak-localization theory with the existence of scattering by some magnetic moments. The fitting parameters demonstrate that disorder most likely comes from the excess iron.

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

1. Introduction

The discovery of iron-based superconductors with superconducting transition temperature up to 56 K has drawn a lot of attention [1–3]. These iron-based superconductors have a common structure, consisting of Fe_2As_2 or Fe_2Se_2 layers. Among these new superconductors, the FeSe_{1-x} system has the simplest structure, with the material consisting of infinitely stacked FeSe layers. The simple structure suggests that FeSe_{1-x} is a good candidate for understanding the origin of superconductivity in the Fe superconductors. FeSe_{1-x} reveals many interesting properties which are different from iron-pnictide superconductors, such as sensitivity to stoichiometry [4], no magnetically ordered state in non-superconducting samples, strong electron-correlation effect [5, 6], and a structural distortion occurring around 100 K without an accompanying magnetic ordering [7]. In addition,

the Se atom in FeSe_{1-x} can be almost replaced by a Te atom, which sustains or even enhances superconductivity [8, 9]. The normal state transport and magnetic properties are modified with Te substitution. The existence of excess iron at interstitial sites in highly Te-substituted $\text{FeSe}_{1-x}\text{Te}_x$ samples has been reported [10–13]. Density functional calculations pointed out that [14] the excess iron at interstitial sites, which provide electronic carriers, are localized and magnetically coupled with the planar Fe. The excess iron can induce the disorder of Fe-derived bands near the Fermi surface. On the other hand, Te substitution has a much weaker influence because the Se/Te-derived band is far away from the Fermi surface [15]. Wen *et al* reported that the magnetic susceptibility of $\text{Fe}_{1+y}\text{Se}_{1-x}\text{Te}_x$, $0.7 \leq x \leq 0.75$, does not follow the Curie–Weiss law above T_C and shows a short-range magnetic ordering at low temperature [16]. In addition, the short-range $(\pi, 0)$ magnetic ordering can be tuned by the

amount of excess Fe or Te [10]. Transport measurements show a logarithmic behavior of resistivity below 50 K in the Te-substituted samples with large amounts of excess iron [17]. This behavior is attributed to the weakly localized state of carriers due to the magnetic coupling between the excess Fe and the Fe at the lattice plane. Similarly, weak localization of charge carriers was observed in $\text{Fe}_{1+y}\text{Se}_{1-x}\text{Te}_x$ samples with high Te concentration ($0.71 \leq x \leq 0.91$) [18]. However, the crossover from metallic to the weakly localized state has not been investigated in detail. Here, we report the properties of magneto-resistivity (MR) accompanied with the resistivity and Hall coefficient of $\text{FeSe}_{1-x}\text{Te}_x$ thin films. The transport properties of pure and slightly Te-substituted $\text{FeSe}_{1-x}\text{Te}_x$ samples show metallic behavior and can be described by a simple two-band model. However, as the Te-substitution level is increased, the quantum interference weak-localization effect becomes significant and results in a negative MR and a semiconductor-like temperature dependence of resistivity. We conclude from these behaviors that the excess iron are responsible for the scattering centers which cause the weak-localization effect.

2. Experimental details

The *c*-axis preferred orientation $\text{FeSe}_{1-x}\text{Te}_x$ thin films were deposited on MgO substrate by the pulsed laser deposition (PLD) technique under vacuum environment ($\sim 10^{-5}$ Torr). Details of sample preparation and characterization were described previously [19]. The x-ray diffraction was performed and the *c*-axis lattice constant was calculated from the (001) peaks using the Nelson–Riley method. Comparing the film data with the results of bulk samples shows the Se concentration was higher than that in the targets. This was consistent with the EDX (energy dispersive x-ray spectroscopy) analysis. However, the EDX analysis cannot give precise results due to the overlapping of the signals of Mg K (from the substrate) and Se L lines. Thus, the Te-substitution level of these thin films was calibrated with the position of the x-ray diffraction peaks (details provided previously [19]). The thin films were patterned into Hall bar geometries for precise measurement by conventional photo-lithography and ion-milling techniques. The width of the main bridge is 50 μm . The transport properties of films including resistivity, Hall voltage, and MR were measured in a Quantum Design physical property measurement system (QD-PPMS).

3. Results and discussion

It has been demonstrated that the iron-based superconductors behave as multi-band systems [20–22]. According to a simple two-band model for non-magnetic samples with lowest order term, the MR due to Lorentz force can be expressed as

$$\text{MR} \equiv \frac{\Delta\rho}{\rho_0} = \frac{(\rho_H - \rho_0)}{\rho_0} \approx \frac{\sigma_h \sigma_e (\mu_h - \mu_e)^2}{(\sigma_h + \sigma_e)^2} B^2 \quad (1)$$

where $\sigma_i = n_i e^2 \tau_i / m_i$ and $\mu_i = e \tau_i / m_i$ (n_i , τ_i , and m_i are carrier density, relaxation time, and effective mass,

Figure 1. Magnetic field dependence of MR for (a) pure FeSe film and (b) $\text{FeSe}_{0.95}\text{Te}_{0.05}$ film at different temperatures. The MR follows a B^2 field dependence. The inset of (a) shows the corresponding Kohler plot, revealing the different temperature dependence of carrier scattering rate.

respectively) are the conductivity and mobility of band *i*, respectively, and ρ_H and ρ_0 are the resistivities of the sample with and without external magnetic field, respectively. The MR due to Lorentz force is positive and has a B^2 field dependence. Figure 1(a) shows the MR of FeSe film and figure 1(b) the MR of $\text{FeSe}_{0.95}\text{Te}_{0.05}$ films under external magnetic fields up to 9 T along the *c*-axis. These MR curves essentially follow a quadratic *B*-field dependence. Interestingly, the coefficient of the B^2 term increases significantly around 100 K in figure 1(a). The coefficient is related to the concentration and scattering rate of carriers, as shown in equation (1). The inset of figure 1(a) shows a Kohler plot, that is MR versus $(B/\rho_0(T))^2$. The slope of the lines has a strong temperature dependence, which violates Kohler's rule [23]. The violation in Kohler's rule indicates that the scattering rate of electron and hole carriers has different temperature dependence. This phenomenon can also be observed in highly Te-substituted samples, which will be shown later. Furthermore, the slopes of MR curves in the Kohler plot increase dramatically around 100 K, the temperature where the structural distortion in the FeSe system occurs. We will discuss this issue later.

Figure 2 presents the temperature evolution of MR curves of $\text{FeSe}_{1-x}\text{Te}_x$ films with various Te-substitution levels. Contrary to the pure FeSe and slightly Te-substituted samples, the negative MR contribution develops gradually and becomes dominant in highly Te-substituted samples. For $x = 0.48$ and 0.56 samples, as shown in figures 2(a) and (b), the competition between these two contributions is observed clearly. As temperature decreases, the MR curve moves toward positive values gradually then turns back toward negative values at lower temperature. This observation indicates that the contribution from negative MR becomes stronger at low temperature. As Te-substitution level further increases, see the $x = 0.85$ sample shown in figure 2(c), the contribution of negative MR dominates in the entire temperature range and the contribution due to Lorentz force becomes negligible.

To obtain a better understanding of the temperature dependence of MR, we plot the MR, resistivity, and Hall coefficient as a function of temperature in figure 3. The contribution from the Lorentz force dominates the value of MR in pure FeSe film in the whole temperature range and the negative MR term becomes stronger as the Te-substitution level increases, as shown in figure 3(a). On the other hand, the ρ - T curve evolves from metallic to semiconductor-like as Te-substitution level increases, figure 3(b). This evolution implies a significant increase of impurity scattering rate of the carriers in highly Te-substituted samples. The weak-localization effect is non-negligible in dirty metals [24–27], which results in a weakly semiconductor-like temperature dependence in resistivity and a negative value in MR. The value of $k_F l$ is a measure to evaluate the contribution of the weak-localization effect, where k_F is the Fermi wave factor and l is the mean free path of carriers. The values of l and v_F can be estimated from the resistivity and Hall coefficient at room temperature using the Drude model. For the $x = 0.85$ sample, the estimated value of $k_F l$ is around 5.8, which indicates a non-negligible contribution of the weak-localization effect in resistivity. In addition, as discussed above, the magnitude of MR increases around 100 K, for both pure FeSe and highly Te-substituted samples. This rise is coincident with the structural distortion of the FeSe superconductor. This significant change is also observed in the Hall coefficient results, as shown in figure 3(c). This phenomenon could result from the change of carrier scattering rate after structural distortion, as discussed previously [28].

The Maki–Thompson correction to MR [29], which results in a positive MR at temperatures close to the superconducting transition temperature, is another fingerprint of the weak-localization effect. To eliminate the positive MR contribution from the Lorentz force, we have measured the MR with external magnetic field parallel to the direction of excitation current. Figures 4(a) and (b) show the magnetic field dependence of MR of $\text{FeSe}_{1-x}\text{Te}_x$ films with $x = 0.48$ and $x = 0.85$, respectively. The MR turns positive gradually as the temperature reaches values close to a few times T_C . In figure 4(c), we present a plot of normalized MR (to the value at 60 K) versus normalized temperature (T/T_C). MR turns positive rapidly as the temperature approaches T_C . It is worth pointing out that the temperature is still

Figure 2. Plots of MR versus magnetic fields along the c -axis at different temperature for $\text{FeSe}_{1-x}\text{Te}_x$ films with $x = 0.48, 0.56$, and 0.85 . The competition between positive and negative MR is observed as temperature is decreased for $x = 0.48$ and 0.56 samples. For the $x = 0.85$ sample, the negative MR contribution dominates in the whole temperature range.

far away from the contribution of paraconductivity resulting from the formation of superconducting pairs above T_C due to thermal fluctuations. Several studies reported similar behavior in dirty superconductors, which were attributed to the Maki–Thompson correction in the weak-localization regime [30, 31].

In general, the inelastic scattering, spin–orbital scattering, Zeeman splitting and weak magnetic impurity scattering have to be taken into consideration [24–27] in the analysis of MR with the weak-localization effect. In addition, the Maki–Thompson correction has to be included for a superconductor as mentioned in the previous paragraph [29–31]. In a three-dimensional system with large electron diffusivities (the diffusion constant $D > 2 \text{ cm}^2\text{s}^{-1}$), with magnetic field perpendicular to the excitation current, the most general form of MR due to weak localization can be expressed as

Figure 3. Temperature dependence of the transport properties of FeSe_{1-x}Te_x films, $x = 0, 0.48, 0.56$, and 0.85 ; (a) MR, (b) resistivity, and (c) Hall coefficient (R_H/ρ^2). The MR is measured under magnetic fields of $+9$ T along the c -axis. The Hall coefficient is calculated from Hall resistivity ρ_{xy} under magnetic fields of $+9$ T and -9 T along the c -axis. The resistivity is obtained without external magnetic field.

$$\frac{\Delta\rho}{\rho^2} = \frac{e^2}{2\pi^2\hbar} \left(\frac{eB}{\hbar} \right)^{1/2} \times \left\{ \left[\frac{1}{2} + \beta(T) \right] f_3 \left(\frac{B}{B_\phi} \right) - \frac{3}{2} f_3 \left(\frac{B}{B_2} \right) \right\}, \quad (2)$$

with $B_\phi = B_{in} + 2B_s$, $B_2 = B_{in} + \frac{2}{3}B_s + \frac{4}{3}B_{so}$, where the characteristic fields $B_x = \frac{\hbar}{4eD\tau_x}$, $\hbar$ is Planck's constant, $x = in, so$, and s , referring to the inelastic, spin-orbit, and magnetic spin-flip scattering times (fields), respectively. Here, $\beta(T)$ is the term responsible for the Maki-Thompson correction. The $f_3(1/x)$ function can be approximated as $f_3(1/x) = 2(\sqrt{2+x} - \sqrt{x}) - (0.5+x)^{-1/2} - (1.5+x)^{-1/2} + (2.03+x)^{-3/2}/48$ within 0.1% accuracy [27]. The magnitude of the spin-orbital field B_{so} is estimated to be around 0.01 T and can be negligible [32]. Furthermore, a Kondo-like scattering due to the magnetic moment based on the s-d exchange model will also give a negative MR with $-B^2$ dependence [33]. Tropeano *et al* have reported a $-B^2$ dependence of MR in highly substituted FeSe_{1-x}Te_x samples $0.8 \leq x \leq 0.9$ [34].

We combine these contributions to analyze our experimental data. Figures 5(a)–(c) show the comparison of MR between experimental data (symbols) and fitting results

(lines) at low temperature. The theoretical curves follow the experimental data with proper choice of fitting parameters (parameters shown in table 1), indicating that the negative MR originates from the weak-localization effect in FeSe_{1-x}Te_x films. Figure 5(d) shows the temperature dependence of β for $x = 0.48$ (squares), $x = 0.56$ (dots), and $x = 0.85$ (triangles) samples. The calculated value (solid line) according to Larkin theory and the data from Cu–Pb films [35] were included for comparison. The Larkin theoretical curve basically follows the value of the β or $x = 0.48$ sample. However, β is significantly suppressed as the Te-substitution level becomes higher.

The fitting parameters are presented in table 1, where α is the coefficient of the $-B^2$ term. First of all, the value of B_{in} decreases gradually at low temperature for all three Te-substituted levels, which indicates a decrease of inelastic scattering of carriers. The inelastic scattering rate most likely arises from the electron–phonon interaction. From the Drude model, we estimate the inelastic scattering rate to be of the order of 10^{12} s^{-1} , about two orders of magnitude smaller than the elastic scattering rate. This result is consistent with the assumption of the domination of impurities in carrier scattering. Second, as the Te-substitution level increases, the electron–magnetic moment scattering (B_s)

Figure 4. The MR as a function of magnetic fields at low temperature for $\text{FeSe}_{1-x}\text{Te}_x$ films with (a) $x = 0.48$ and (b) 0.85. The value of MR becomes positive gradually as the temperature approaches T_c . (c) MR normalized to the value at 60 K as a function of reduced temperature T/T_c , for $x = 0.48, 0.56$, and 0.85 films. The MR is measured under a magnetic field of 9 T parallel to the direction of excitation current. The emergence of positive MR is suppressed to a lower temperature in the higher Te-substituted sample.

Figure 5. The comparison of MR at low temperatures between experimental data (symbols) and the fitting curves (lines) for different Te-substitution levels, (a)–(c). The fitting parameters are shown in table 1. The experimental data can be fitted well for all temperatures. The temperature dependence of the fitted Maki–Thompson coefficient, β , is shown in (d). The solid line is the calculated value according to Larkin theory and the results from Cu–Pb films are also included for comparison [35]. The value of β is suppressed significantly in highly Te-substituted samples.

becomes non-negligible; the Maki–Thompson correction (β) is strongly suppressed; and the contribution of Kondo-like carrier–magnetic moment interaction (α) becomes slightly

stronger. We believe these to be evidence for more impurities having localized magnetic moments in highly Te-substituted samples.

Table 1. Fitting parameters of MR for higher Te-substituted $\text{FeSe}_{1-x}\text{Te}_x$ films at low temperature. The contribution of spin–orbital interaction and Lorentz force are neglected. Here, B_{in} , B_{S} , and β are defined in the text, and α is the coefficient of the $-B^2$ term from the Kondo-like scattering due to the magnetic moment based on the s–d exchange model.

Te-level x	Temp. (K)	B_{in} (T)	B_{S} (T)	β	α
0.48	80	5.6	0.73	0	0
	60	4.3	0.73	0	0
	40	2.7	0.73	0.5	0
	20	1.4	0.73	2.7	0
0.56	80	7	0.85	0	0
	50	3.8	0.85	0	0.05
	20	1.8	0.85	0.8	0.1
0.85	80	4.9	1.0	0	0
	60	3.5	1.0	0	0.08
	40	2.9	1.0	0	0.28
	20	1.8	1.0	0.05	0.35

Combining the MR, resistivity, and Hall coefficient results, the effect of Te substitution in FeSe_{1-x} superconductors can be understood as follows. Scattering of carriers due to disorder potential is significantly enhanced as Te is substituted, which makes the weak-localization effect non-negligible. This effect becomes stronger as the Te-substitution level is higher. Hence, the resistivity versus temperature curves for $\text{FeSe}_{1-x}\text{Te}_x$ evolve from metallic to semiconductor-like behavior and the MR changes from a positive value due to cyclotron motion of carriers to a negative value due to weak-localization effects. The structural distortion seems to have a different effect on the scattering rate of hole and electron carriers, resulting in a strong temperature dependence of Hall coefficient and MR. Recently, Sood *et al* [36] reported the significant hardening of S1 mode from Raman scattering results. This might be an important clue to understanding the effect of structural distortion on the scattering rate of carriers.

What is responsible for these disordered potentials? Neglecting crystalline defects, two possible disorder potentials could be formed when Te is substituted at the Se position. First, the substituted Te atoms randomly distribute at the Se sites because no phase separation is observed in their x-ray diffraction patterns. This random distribution of Te/Se atoms in the lattice provides possible disordered potentials. Another possible source for disordered potentials is the excess Fe at the interstitial sites, as pointed out in neutron experiments [10–13]. According to density functional theory (DFT) calculations [14], the excess Fe atoms perturb the Fe-derived bands near the Fermi surface. However, the random distribution of Te/Se mainly perturbs the Se-derived bands away from the Fermi surface. Our experimental results demonstrate that the strength of the weak-localization effect is stronger in the higher Te-substituted sample, rather than in a sample with Te/Se ratio close to 1. We believe that the carriers will be influenced more effectively by excess Fe atoms. In addition, the excess Fe atoms probably have a localized magnetic moment [10, 14, 18], which is consistent with our fitting results. Therefore, our results strongly indicate

that the excess iron is responsible for the disordered potentials in Te-substituted $\text{FeSe}_{1-x}\text{Te}_x$ superconductors.

4. Conclusion

In conclusion, the MR properties of $\text{FeSe}_{1-x}\text{Te}_x$ superconducting thin films have been investigated. In contrast to the positive MR observed in FeAs superconductors [37–40], a negative MR was observed in Te-substituted $\text{FeSe}_{1-x}\text{Te}_x$. For pure FeSe and slightly Te-substituted $\text{FeSe}_{1-x}\text{Te}_x$ samples, the MR is dominated by the cyclotron motion of carriers due to the Lorentz force, and shows positive quadratic magnetic field dependence. As Te is substituted for Se, the weak-localization effect becomes non-negligible and results in a negative MR. The negative MR with a $-B^2$ field dependence was also observed in highly substituted $\text{FeSe}_{1-x}\text{Te}_x$ samples $0.8 \leq x \leq 0.9$, and this observation was attributed to Kondo-like electron–moment interaction [34]. Our result suggests the necessary consideration of the weak-localization effect. The MR at low temperature can be well described by combining the contributions from weak-localization theory with the Maki–Thompson corrections and from Kondo-like electron–moment interaction. In addition, by analyzing the evolution of MR with Te substitution, we believe the disorder arises mainly from the excess iron. These results allude to the important role excess iron plays in the 11 series iron-based superconductors.

Acknowledgments

This work was supported by the National Science Council, Taiwan, grant nos 98-2119-M-001-025 and 99-2112-M-001-028-MY3.

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