Transport properties of FeSe epitaxial thin films under in-plane strain

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Abstract. We demonstrate successful fabrication of FeSe films with various magnitude of lattice strain, from tensile to compressive. The superconducting transition temperature, $T_c$, shows the systematic change by in-plane strain. It reaches 12 K for the films with compressive strain whereas the superconductivity disappears for the films with large tensile strain. The structural transition temperature, $T_s$, decreases slightly as the in-plane strain is more compressive. In other words, there is a negative correlation between $T_c$ and $T_s$. It is also found that both the hole and the electron carrier densities evaluated by transport properties increase as the in-plane strain is more compressive. Therefore, there is a clear correlation between $T_c$ and the carrier densities, which suggests that it is essential for the $T_c$ enhancement of iron-based superconductors to increase carrier densities.

1. Introduction

Discovery of iron-based superconductors (FeSCs) has attracted much attention to both fundamental studies and applications[1]. An iron-chalcogenide superconductor, FeSe[2], is one of the most suitable material for understanding the superconductivity of the FeSCs because it has the simplest crystal structure among the FeSCs, composed of conducting planes alone. The superconducting transition temperature, $T_c$, of the bulk crystal FeSe at ambient pressure is 9 K. However, it is increased up to 38.3 K under high pressure[3, 4, 5]. In single-layer FeSe films on SrTiO$_3$ substrates, $T_c$ increased dramatically up to about 65 K[6, 7] or even higher[8]. From these point of view, FeSe has the high-$T_c$ potential. Therefore, it is important to reveal the superconductivity mechanism of FeSe.

As was demonstrated by the pressure effect[3, 4, 5], $T_c$ of FeSe is sensitive to the lattice strain. Previously, we succeeded in obtaining FeSe$_{1-x}$Te$_x$ thin films for all $x$ values, which is impossible in bulk crystal, and $T_c$ of these films reaches 23 K at $x = 0.2$[9]. Since Te-substitution introduces a negative chemical pressure to FeSe, this result shows that $T_c$ of FeSe was strongly affected by the isotropic pressure. On the other hand, even in the samples of FeSe$_{1-x}$Te$_x$ with the same $x$ value, there is a negative correlation between the $a$-axis and $c$-axis parameters[9, 10, 11], suggesting that the anisotropic strain may play some role. Since the essential feature of the $T_c$ enhancement in FeSe films is that the films are compressed along the $a$-axis[12, 13], the separation of the effect of the anisotropic pressure from the isotropic pressure is important for the correct understanding of the pressure effect of iron-chalcogenides. Thus, we focused on the role of the anisotropic strain, and prepared FeSe films with various magnitude of in-plane strain, including both compressive and tensile, where the transport properties were investigated.
2. Method

FeSe films were grown under vacuum (∼10⁻⁸ Torr) by the pulse laser deposition (PLD) method using a KrF laser[15, 16]. An Fe₁₂Se polycrystalline pellet was used as a target. The single-crystalline substrates used in this study were LaAlO₃ (LAO, a=3.789 Å) and (LaAl)₀.₇₋(SrAl₀.₅Ta₀.₅)₀.₃O₃ (LSAT, (1/2)a=3.868 Å). We also prepared FeSe on LSAT with LAO buffer, to control the magnitude of strain. The a-axis and the c-axis lengths of the films were determined from the (204) and (00l) reflections in the X-ray diffraction (XRD) measurements. The thicknesses of the grown films were 30~75 nm, measured by a Dektak 6 M stylus profilometer. For transport measurements, these films were deposited in a six-terminal shape using a metal mask. The measured area was 1.2 mm long and 1.0 mm wide. The electrical resistivity and the Hall effect were measured using Physical Property Measurement System (PPMS, Quantum Design, Inc.) from 2 to 300 K.

3. Result and Discussion

Figure 1(a)-(c) show the XRD patterns of the grown FeSe thin films on the three different substrates. These films showed only the (00l) reflections of the tetragonal PbO structure except for peaks from Ag paste, which indicates that the films were well-oriented along the c-axis. A four-fold symmetry of the phi-scan of the (101) reflection was also confirmed. Figure 1(d) shows the enlarged plots of Fig. 1(c) around the (001) peak. The Laue fringe is clearly observed and the intensity of each peak also well agrees with theoretically expected ones, which demonstrate that the grown films have flat surface. Figure 1(e) shows the relation between the a-axis and the c-axis lengths of the films, which shows both a and c parameters change continuously for a wide
Figure 2. (a) Temperature dependence of the electrical resistivity, \( \rho \), of the FeSe thin films and (b) the enlarged plots of (a) around \( T_c \).

range with the negative correlation. The fact that the films with longer \( a \) has shorter \( c \) can be understood simply by the Poisson effect. Therefore, all of those demonstrate that we succeeded in obtaining a series of single-crystalline films with various magnitude of anisotropic strain. The in-plane parameter, \( \varepsilon = (a - a_{\text{bulk}})/a_{\text{bulk}} \), is varied from -1.0% to +1.5% in this study.

Figures 2(a) and (b) show the temperature dependence of the electrical resistivity, \( \rho \). The “residual” resistivity ratio, \( \text{RRR} = \rho(300 \text{ K})/\rho(0 \text{ K}) \), reaches \(~18\), which shows that our films have good crystalline quality. Note that a kink anomaly which corresponds to the structure transition is observed at about 90 K in the \( \rho-T \) curve as bulk FeSe\[17\]. The structural transition temperature, \( T_s \), can be determined as a position of the kink anomaly in the \( \text{d}\rho/\text{dT-T} \) curve, and the in-plane strain dependence of \( T_s \) is discussed soon later. It is noteworthy that \( T_c \) changes systematically along with the change of the in-plane strain parameter, \( \varepsilon \). \( T_c \) reaches 12 K for the films with compressive strain whereas the superconductivity disappears for the films with large tensile strain.

Figure 3 shows the temperature dependence of the Hall coefficient, \( R_H \), of the films. In each film, the behavior above 100 K are similar. \( R_H \) is rather small, and with decreasing temperature, \( R_H \) changes from negative to positive below 120 K. On the other hand, with further decreasing temperature, \( R_H \) starts to depend on \( \varepsilon \). As \( \varepsilon \) becomes larger, \( R_H \) become larger at low temperatures.

Figures 4(a)-(c) show \( T_s \), \( T_c \) and \( R_H \) at 20 K as a function of \( \varepsilon \), respectively. As was already described, when \( \varepsilon \) increases, \( T_c \) decreases systematically and disappears rapidly in the films with the large tensile strain. On the other hand, \( T_s \) does not strongly depend on \( \varepsilon \), but slightly decreases as \( \varepsilon \) becomes smaller. In other words, \( T_c \) and \( T_s \) has a negative correlation. This negative correlation is consistent with the sudden increase of \( T_c \) at the disappearance of the structural transition in Te-substituted films[11]. Since the structural transition is thought to be the so-called nematic transition, this negative correlation between \( T_c \) and \( T_s \) suggests that the
Figure 3. Temperature dependences of Hall coefficients, $R_H$, of the FeSe films. A result of bulk FeSe[18] and FeSe film on CaF$_2$[12] are also plotted.

Figure 4. The in-plane strain parameter ($\varepsilon$) dependence of (a) $T_s$, (b) $T_c$, and (c) $R_H$ at 20 K. A result of bulk FeSe[18] are also plotted. The hatched curves are guides for the eye.
Figure 5. The in-plane strain parameter, $\varepsilon$, dependence of (a) carrier density and (b) mobility. The dashed curves are guides for the eye.

electronic nematicity is unfavorable to raise $T_c$ in iron-chalcogenide.

The systematic change of $R_H(20 \text{ K})$ as a function of $\varepsilon$ in our films was also observed. Note that $R_H(20 \text{ K})$ of the bulk FeSe[18] does not fit to our result in films. This difference may be caused by the difference of the chemical composition or the difference in the local structural parameters such as bond angle and anion height between films and bulk crystal. In any case, we believe that this difference in the low-$T$ $R_H$ behavior between films and bulk singles crystal is not important for superconductivity because $T_c$ of the bulk crystal is fitted to our data in films.

In multiple-band systems like iron-chalcogenides, $R_H$ is not related to the carrier density in a simple form. To discuss the $R_H$ data in more detail, we apply a text-book approach for multiple-band materials. In order to evaluate the carrier density and the mobility of the films, we also measured the magneto-resistance. In a classical two-band model, the resistivity tensor is expressed as

$$\rho_{xx}(0) = \frac{1}{e(n_e \mu_e + n_h \mu_h)}, \quad (1)$$

$$\rho_{yx}(B) = \frac{-n_e \mu_e^2 + n_h \mu_h^2}{e(n_e \mu_e + n_h \mu_h)^2} B, \quad (2)$$

$$\rho_{xx}(B) = \frac{n_e n_h \mu_e \mu_h (\mu_e + \mu_h)^2}{(n_e \mu_e + n_h \mu_h)^2} B^2, \quad (3)$$

where $\mu_h$, $\mu_e$, $n_h$, and $n_e$ are the hole mobility, the electron mobility, the hole density and the electron density, respectively. Because there are four parameters for three equations, we assumed the carrier density ratio ($n_h/n_e$) based on an angle-resolved photoemission spectroscopy (ARPES) data[19].

The obtained $\mu_h$, $\mu_e$, $n_h$ and $n_e$ are summarized in Fig. 5. Both the hole and the electron carrier densities increase systematically as $\varepsilon$ becomes smaller. Recently, the ARPES study using our films also showed the increase of the hole and the electron densities in an in-plane compressed sample[19]. This agreement between our Hall data and the ARPES study demonstrate the validity of the conclusion we obtained. Namely, the increase of the carrier densities is essential
for the increase of $T_c$. On the other hand, both the hole and the electron mobility are almost unchanged in the films on oxide substrates. In addition, those of FeSe on CaF$_2$ is rather small. In the FeSe films on CaF$_2$, TEM observation showed that F$^-$ ions penetrated into the films from the CaF$_2$ substrates[20, 21, 22], which may be the reason for small mobilities of FeSe on CaF$_2$. The fact that $T_c$ is the highest in films with the small mobilities suggests that the mobility is not important for superconductivity. Rather, our result shows that the most essential factor to realize high $T_c$ is the increase of the carrier densities in iron-chalcogenides.

4. Conclusion

We succeeded in fabricating a series of FeSe films with various magnitude of the in-plane strain, including both tensile and compressive. $T_c$ and $R_H$ show the systematic change by in-plane strain, $\varepsilon$, whereas $T_s$ does not strongly depend on $\varepsilon$ and decreases slightly as $\varepsilon$ becomes smaller. This negative correlation between $T_c$ and $T_s$ suggest that the electronic nematicity is unfavorable to raise $T_c$. The carrier densities evaluated by transport properties also increase systematically as $\varepsilon$ becomes smaller. We demonstrate a clear correlation between $T_c$ and the carrier densities, which suggests that it is essential for the $T_c$ enhancement of iron-chalcogenides to increase the carrier densities.

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