Iron-based superconductors (FeBSs) are attracting widespread interest in the field of superconductivity in terms of both fundamental and applied research. An iron chalcogenide superconductor, FeSe$_{1-x}$Te$_x$ (1,2) is a FeBS with the simplest crystal structure, consisting of conducting planes alone. The parent material FeSe shows a structural transition from a tetragonal to an orthorhombic phase at 90 K,3 while it exhibits no magnetic order at ambient pressure, different from many other parent materials of FeBSs. It is considered that the structural transition has an electronic origin,4,5 and is also called the nematic transition. FeSe exhibits superconductivity below 9 K. The Te substitution of Se increases $T_c$ of 14 K is obtained at $x=0.5$ at ambient pressure.6 In spite of its low $T_c$ compared with other iron-based superconductors, it maintains large critical current densities, $J_c$, at very high magnetic fields,6-9 and thus, FeSe$_{1-x}$Te$_x$ has great potential in high-field applications such as superconducting wires and tapes for magnets.

Much research on the thin-film growth of FeSe$_{1-x}$Te$_x$ has focused on the composition of $x=0.5$ — the optimal composition for achieving high $T_c$ in bulk samples — and enhanced superconductivity due to the in-plane lattice strain ($T_c^{x=0.5} \approx 19$ K at the maximum)9 has been reported.6,7,9,10 However, the optimal composition of film samples is different from that of bulk samples. Although bulk samples with $0.1 < x < 0.4$ of FeSe$_{1-x}$Te$_x$ are not available because of phase separation, thin-film growth techniques make it possible to obtain single crystalline samples of FeSe$_{1-x}$Te$_x$ in the whole composition region.11-13 The true optimal composition of film samples is in the phase separation region of $0.1 < x < 0.4$.

We previously grew FeSe$_{1-x}$Te$_x$ films on two different substrates, namely, CaF$_2$ and LaAlO$_3$ (LAO), and have obtained two electronic phase diagrams for these samples.12,13 Although the detailed behaviors of the two phase diagrams are different from each other, they show the same behavior qualitatively: (i) Te substitution decreases the nematic (structural) transition temperature, $T_n$, (ii) when the nematic transition disappears, $T_c$ rapidly increases, and the highest $T_c$ is obtained just after the disappearance of the nematic transition, and (iii) $T_c$ decreases monotonically with increasing $x$ in the tetragonal phase. The compositions, $x_c$, where the structural transition disappears are different between the two phase diagrams; $x_c$ is smaller for films on CaF$_2$ ($x_c=0.2$ for CaF$_2$ and $x_c=0.4$ for LAO).

The fact that the sudden increase in $T_c$ at $x_c$ is commonly observed in the two different phase diagrams strongly indicate that the nematicity suppresses the superconductivity in FeSe$_{1-x}$Te$_x$. Because the maximum value of $T_c$ is higher for films on CaF$_2$ than on LAO, we expect the maximum value of $T_c$ to become larger if we can suppress the nematicity faster. Therefore, it is essential to elucidate the origin of the difference in $x_c$ between films on CaF$_2$ and on LAO for further enhancement of superconductivity in FeSe$_{1-x}$Te$_x$. From a simplistic point of view, the origin of the difference in the phase diagrams between films on the two substrates is expected to be in the difference in the strength of the lattice strain. Indeed, there is a tendency that the $a$-axis length of films on CaF$_2$ is shorter than that on LAO. Therefore, it is important to investigate the effects of strain on the physical properties of these materials.

In this letter, we report on the systematic studies of transport properties of FeSe thin films with various degrees of strain. We demonstrate that the structural transition is suppressed by compressive strain, consistent with the fact that the suppression of $T_c$ is stronger in films on CaF$_2$ than on LAO. This result suggests that stronger compressive strain can make $x_c$ smaller, which may result in further enhancement of $T_c$ in FeSe$_{1-x}$Te$_x$ thin films. In addition, we report a clear correlation between $T_c$ and the carrier concentrations, which suggests that it is essential to increase carrier densities for realizing high $T_c$ in FeSe. Our results will provide important clues for further enhancement of $T_c$ as well as the superconducting mechanism in iron chalcogenides.

All the FeSe thin films were grown by a pulsed laser deposition method using a KrF laser.14,15 To change the strength of in-plane strain we grew films on three different substrates in this study, namely, LaAlO$_3$ (LAO), (LaAl)$_{0.7}$–(SrAl)$_{0.3}$TaO$_3$ (LSAT), and LSAT with a LAO buffer layer. The purpose of the use of the LAO buffer layers on LSAT is to avoid possible diffusion of oxygen at the interface of film and LSAT substrates. However, it turned out that there were no significant differences in the crystalline quality and the superconducting properties between films on LSAT with and without LAO buffer layer. The crystal structures and the orientations of the films were characterized with a four-circle X-ray diffractometer with Cu Kα radiation at room temperature. The thicknesses of the samples were evaluated with a Dektak 6M stylus profiler. The electrical resistivity and the Hall resistivity were measured using a physical property measurement system from 2 to 300 K under magnetic fields up to 9 T.

Figure 1(a) shows the X-ray diffraction (XRD) patterns of three typical samples on different types of substrates. Except...
The transition temperature, $T_s$, can be determined from the position of the superconducting transition. Note that a kink anomaly which is due to the structural transition was observed at around 90 K in the bulk samples. The structural transition increases with increasing compressive strain parameter, $\varepsilon > 1.5\%$.

Figure 2(a) shows the temperature dependence of the dc electrical resistivity, $\rho$, of FeSe films with various strain. The temperature dependence of $d\rho/dT$ of a film with $\varepsilon \sim +0.4\%$ is also plotted. (b) Temperature dependence of the resistivity of FeSe films around the superconducting transition temperatures.

Figure 3 shows $T_c$ and $T_s$ as a function of the in-plane strain parameter, $\varepsilon$. As already described, when $\varepsilon$ increases from negative to positive, $T_c$ decreases systematically. $T_c$ drops rapidly in tensile-strained films and films with $\varepsilon > 0.5\%$ do not show zero resistivity above 2 K. A recent angle-resolved photoemission spectroscopy (ARPES) study indicates that the rapid decrease in $T_c$ for films with tensile strain is related to a Lifshitz transition. On the other hand, $T_s$ increases with increasing $\varepsilon$. In other words, there is a negative correlation between $T_s$ and $T_c$ in FeSe under in-plane strain. This may suggest that the electronic nematicity is unfavorable for raising $T_c$ in iron chalcogenides, consistent with the sudden increase in $T_c$ at the disappearance of the nematic transition in Te-substituted films.

The decrease in $T_c$ due to compressive strain observed for the FeSe films suggests that the structural transition can be extinguished by a smaller amount of Te substitution for films with more compressive strain. This is consistent with the previous results in our FeSe$_{1-x}$Te$_x$ films that the $T_c$ of films on...
CaF$_2$ is smaller than that of films on LAO, which have longer $a$-axis lengths than films on CaF$_2$. As described earlier, considering the fact that (i) the maximum of $T_c$ is obtained for films with $x \approx x_c$ and (ii) $T_c$ increase with decreasing $x$ for $x > x_c$, it is expected that the realization of smaller $x_c$ would lead to higher $T_c$. Our results indicate that this is possible by applying more compressive stress in FeSe$_{1-x}$Te$_x$.

To reveal the nature of charge carriers in the grown films we performed Hall measurements as well as magneto-resistance measurements. Figure 4(a) shows the temperature dependence of the Hall coefficient, $R_H$, of the grown films. The behavior of $R_H(T)$ above 100 K is very similar among the samples, including bulk crystals, and the values of $R_H$ are very small. On the other hand, $R_H$ increases significantly with decreasing temperature below 100 K, and the strain dependence becomes visible. $R_H$ becomes large with increasing $\varepsilon$ at low temperatures. Note that the $R_H(T)$ of bulk samples deviates from those of films below 100 K, which decreases on cooling below 70–80 K and becomes negative at low temperatures.\textsuperscript{23} We will discuss the origins of the difference in the low-$T\ R_H$ behavior between films and bulk later.

In a multiband system like iron chalcogenides, where electron- and hole-type carriers coexist, $R_H$ is not related to the carrier densities in a simple form. We considered one electron band and one hole band representing the multiple bands and applied the text-book approach for multiband materials. In a classical two-band model, the resistivity tensor is expressed as

$$\rho_{xx}(0) = \frac{1}{e(n_{\text{h}}\mu_{\text{h}} + n_{\text{e}}\mu_{\text{e}})},$$

$$\rho_{xx}(B) - \rho_{xx}(0) = \frac{n_{\text{h}}n_{\text{e}}\mu_{\text{h}}\mu_{\text{e}}(\mu_{\text{h}} + \mu_{\text{e}})}{(n_{\text{h}}\mu_{\text{h}} + n_{\text{e}}\mu_{\text{e}})^2} B^2,$$

$$\rho_{xy}(B) = \frac{n_{\text{h}}\mu_{\text{h}}^2 - n_{\text{e}}\mu_{\text{e}}^2}{e(n_{\text{h}}\mu_{\text{h}} + n_{\text{e}}\mu_{\text{e}})^2} B,$$

where $n_{\text{h}}$, $n_{\text{e}}$, $\mu_{\text{h}}$, and $\mu_{\text{e}}$ are the hole density, the electron density, the hole mobility, and the electron mobility, respectively. We evaluated the carrier densities and mobilities of the films with the measured data of $R_H$ and the magnetoresistance, assuming $n_{\text{h}} = n_{\text{e}}$.\textsuperscript{22}

The obtained values of carrier densities and mobilities are plotted as a function of $\varepsilon$ in Fig. 4(b). As $\varepsilon$ decreases, the carrier densities increase. This result is consistent with the ARCES results, which also showed the increase in both hole and electron densities for an in-plane compressed sample.\textsuperscript{20} The agreement between our results of the transport measurements and the ARCES study demonstrates that there is a correlation between the $T_c$ and the carrier densities of FeSe. This suggests that the increase in the carrier densities is essential for the increase in $T_c$.

On the other hand, we found no significant correlation between both the hole and electron mobilities and $\varepsilon$. The fact that the highest $T_c$ is obtained for films with small mobilities implies that the mobility is not so important for superconductivity in FeSe. Note that the hole mobility is always higher than that of electron for our films. However the opposite behavior was reported for bulk FeSe,\textsuperscript{23} which results in the difference in the sign of $R_H$ at low temperatures between films and bulk samples. Although the origin of this difference in mobilities between films and bulk single crystals is unclear at present, we believe that it is not important for superconductivity because the $T_c$ of the bulk crystal does not significantly deviate from the $T_c$-vs-$\varepsilon$ curve for our films. Indeed, a mobility spectrum analysis\textsuperscript{23} revealed that bulk FeSe has minority N-type carriers with very high mobility, while majority of both N- and P-type carriers have comparable mobilities, which may result in higher mobility of electron-like carriers in the two-band model. These minority carriers with high mobilities are considered to contribute insignificantly to $T_c$.

Finally, we comment on the relationship between the superconductivity and the nematicity in iron chalcogenides. Our results with strained FeSe are that $T_c$ decreases and $T_c$ increases by applying compressive stress. This is very similar to the behavior under hydrostatic pressures below approximately 1 GPa.\textsuperscript{24} These results with strained FeSe may suggest that the electronic nematicity is unfavorable for raising $T_c$ in iron chalcogenides. As described earlier, this is consistent with the results for the Te-substituted samples, where a rapid increase in $T_c$ is observed corresponding to the disappearance of the nematic transition.\textsuperscript{31} Another isovalent substitution by sulfur also suppresses the nematicity. However, there is no significant increase in $T_c$ when the nematicity disappears, or rather $T_c$ decreases after the disappearance of the nematicity for S-substituted samples.\textsuperscript{25–28} The contrasting phase diagrams of FeSe$_{1-x}$Te$_x$ and FeSe$_{1-x}$S$_x$ indicate that the role of the nematicity is not universal in the superconductivity of iron chalcogenides, suggesting that the nematicity affects $T_c$ only in an indirect manner.\textsuperscript{28} Rather, our results suggest that the most essential factor for realizing high $T_c$ is the increase in the carrier densities.

This conclusion may be inconsistent with our previous results of Hall measurements with FeSe$_{1-x}$Te$_x$ films,\textsuperscript{29–31} which suggested that the $n_{\text{h}}$-to-$n_{\text{e}}$ and/or $\mu_{\text{h}}$-to-$\mu_{\text{e}}$ ratio were essential for high $T_c$. This disagreement may suggest that there are multiple channels for increasing $T_c$, originating from the multiband/multiorbital character in iron chalcogenides.\textsuperscript{32} In other words, it is necessary to increase carrier densities for...
obtaining high \(T_c\) of up to 12 K in FeSe, and for further enhancement of \(T_c\) of up to 23 K in FeSe\(_{1-x}\)Te\(_x\), we may need to tune the ratio of carrier densities and/or mobilities. Further comprehensive and systematic studies with Te- and S-substituted samples are needed for a complete understanding of the behavior of \(T_c\) in iron chalcogenides, which is now under way.

In conclusion, we have succeeded in growing a series of FeSe films with various degrees of the in-plane strain, from tensile to compressive. We found that as the strain becomes more compressive, the structural transition temperature decreases. This result suggests that the difference of the substitution content \(x\) that is required for the complete suppression of the structural transition between FeSe\(_{1-x}\)Te\(_x\) films on LAO and on CaF\(_2\) is due to the difference in the degree of strain. This means that the structural transition can be extinguished by a smaller amount of Te substitution for films with more compressed strain, which may lead to higher \(T_c\). It was also found that \(T_c\) and the carrier densities of the FeSe films increase systematically as \(\varepsilon\) decreases. The clear correlation between \(T_c\) and the carrier densities suggests that for the \(T_c\) enhancement of iron chalcogenides it is essential to increase the carrier densities.

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