

# Superconductivity under Pressure in $\text{FeSe}_{1-x}\text{Te}_x$ Studied by DC Magnetic Measurements

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## Abstract

Superconductivity under pressure in  $\text{FeSe}_{1-x}\text{Te}_x$  with  $x=0.7$  ( $T_c \sim 14$  K) has been investigated by the measurements of DC magnetization using high quality single crystal specimens. It has been found that  $T_c$  increases and makes a maximum of  $\sim 15$  K at 1 GPa but rapidly decreases above 1 GPa under hydrostatic pressure using liquid Ar as pressure transmitting media (PTM). In contrast,  $T_c$  is found to increase up to 18 K at 2.5 GPa then decrease gradually under nearly uniaxial pressure along  $c$ -axis using NaCl as PTM. It is also found that  $T_c$  reaches a maximum of 16 K at 1 GPa but is nearly pressure independent above 1 GPa under nearly uniaxial pressure along  $a$ -axis. These behaviors suggest that the superconductivity is suppressed by the isotropic compression but is enhanced (not changed) by the uniaxial compression along  $c$ -axis ( $a$ -axis).

*Keywords:* superconductivity, iron-pnictides,  $\text{FeSe}_{1-x}\text{Te}_x$ , high pressure

## 1 Introduction

Discovery of superconductivity in  $\text{LaFeAsO}_{1-x}\text{F}_x$  ( $T_c=26$  K)[1] has attracted much interest in understanding the pairing mechanism and also in searching new related iron-pnictide superconductor, where superconductivity occurs in the iron-pnictide (Fe-Pn) layers consisting of edge-sharing  $\text{FePn}_4$  tetrahedron. It has been pointed that pnictogen height  $h_{\text{Pn}}$  and Pn-Fe-Pn bond angle, in addition to the lattice constant, are the important structural parameters to determine  $T_c$ [2, 3]. Thus, to elucidate the mechanism of superconductivity, it is important to clarify the relation between these parameters and  $T_c$  by investigating the relation under physical and chemical pressure, as demonstrated in  $\text{RFeAsO}_{1-x}\text{F}_x$  (R=La, Ce-Sm)[4].

$\text{FeSe}_{0.3}\text{Te}_{0.7}$  ( $T_c=14$  K) is an iron-pnictide superconductor with the simplest tetragonal crystal structure, which is composed only by Fe-(Se, Te) layers stacked along  $c$ -axis. In the end material FeSe ( $T_c=8$  K),  $T_c$  has been found to be enhanced up to 34 K under hydrostatic pressure mainly due to the change of the crystal structure from orthorhombic to tetragonal one under pressure[5, 6]. On the other hand, investigations on the superconductivity under pressure in  $\text{FeSe}_{1-x}\text{Te}_x$  have been limited to the polycrystalline specimens[7], which shows non bulk superconductivity with slightly lower  $T_c$  due to the excess Fe in the interstitial sites of (Te, Se)

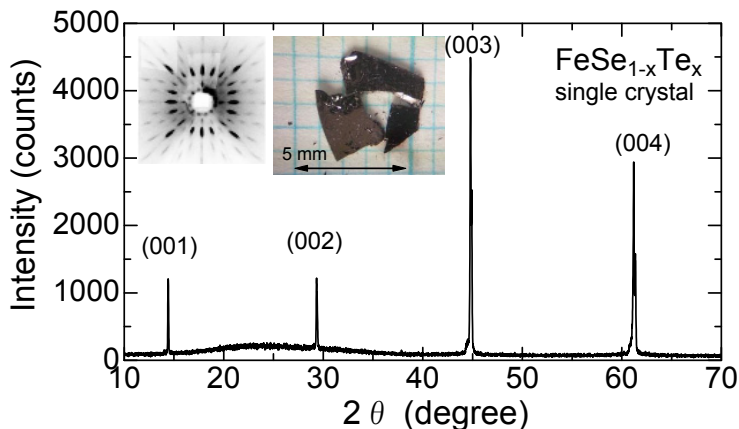


Figure 1: X-ray diffraction pattern of  $\text{FeSe}_{1-x}\text{Te}_x$  single crystals at room temperature. Diffraction peaks from  $(00l)$  planes are observed. The insets show X-ray Laue pattern observed along  $c$ -axis and a photograph of  $\text{FeSe}_{1-x}\text{Te}_x$  single crystals.

layers[8, 9]. Recently, high quality single crystal specimens which shows bulk superconductivity have been produced by annealing crystals in Te vapor or vacuum after the single crystal growth by Bridgman method[10, 11, 12, 13, 14]. Thus, further study on the superconductivity under pressure should be done by using high quality single crystal specimens to clarify the intrinsic pressure evolutions of  $T_c$  precisely. In the present work, we have performed DC magnetization measurements under hydrostatic and nearly uniaxial pressure using single crystal specimens. Our DC magnetic measurements under pressure is a powerful technique to reveal intrinsic  $T_c$ , as successfully applied to other superconductors[4, 5, 15, 16].

## 2 Experimental

Single crystal specimens with nominal composition  $\text{FeSe}_{0.3}\text{Te}_{0.7}$  were grown by Bridgeman method in a similar manner as described in the literatures[9, 10, 11, 12, 13, 14]. First, an appropriate amount of Fe, Se, and Te was mixed thoroughly in an Ar atmosphere. The mixture was loaded and sealed in an evacuated silica tube. The silica tube was heated to 600 °C for 24 h and heated 1000 °C 12 h, and then cooled slowly to 650 °C at a rate of 3.0 °C/h. Small pieces of the as-grown crystals and Te were sealed in a evacuated silica tube, where they were put at both ends of the tube. The silica tube was heated 400 °C 24 h, followed by water quenching. Phase purity of the single crystals was checked by powder x-ray diffraction. Figure 1 shows X-ray diffraction pattern of  $\text{FeSe}_{1-x}\text{Te}_x$  single crystals, indicating that  $(00l)$  plane is exposed on the crystal surface. DC magnetization measurements under pressure were done by using a miniature diamond anvil cell (DAC) combined with a sample rod of a commercial SQUID magnetometer. Details of the DAC are given in elsewhere[17]. A single crystal specimen was loaded into the gasket hole together with a small piece of high-purity lead (Pb) to realize the in situ observation of pressure by determining the pressure from the  $T_c$  shift of Pb. As pressure transmitting media (PTM), we used liquid Ar and NaCl to generate hydrostatic and nearly uniaxial pressure, respectively. Liquid Ar is known to generate high pressure with good hydrostaticity up to 10 GPa[18].

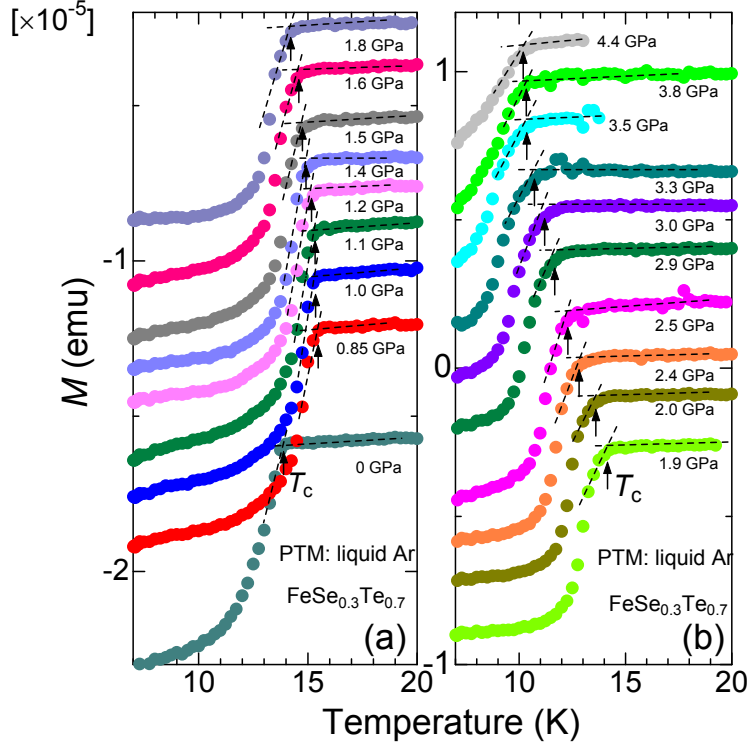


Figure 2: Temperature dependence of zero-field-cooled DC magnetization for  $\text{FeSe}_{1-x}\text{Te}_x$  ( $x=0.7$ ) measured in a magnetic field of 20 Oe under various pressures below 1.8 GPa (a) and above 1.9 GPa (b). The measurements have been done by using liquid Ar as a pressure transmitting medium (PTM). The data are intentionally shifted along the longitudinal axis for clarity.

### 3 Results and Discussion

We show the temperature dependence of the zero-field-cooled DC magnetization ( $M$ ) for  $\text{FeSe}_{1-x}\text{Te}_x$  ( $x=0.7$ ) under various pressures up to 1.8 GPa in Fig. 2(a) and higher pressure up to 4.4 GPa in Fig. 2(b) measured using liquid Ar as PTM to compress the single crystals under hydrostatic condition. We define the onset of the diamagnetic response, which is estimated by extrapolating the initial slope of the  $M$ - $T$  curve just below the decrease to the normal state magnetization, as  $T_c$ . As seen in Fig. 2(a), the  $M$ - $T$  curve at ambient pressure ( $P=0$  GPa) shows a rapid decrease below  $T_c \sim 14$  K. At  $P=0.85$ -1.0 GPa, the diamagnetic onset shifts to higher temperature side, indicating  $T_c \sim 15$  K. Above 1.0 GPa, the diamagnetic onset shifts to lower temperature side monotonically as increasing pressure down to 14 K at 1.8 GPa in Fig. 1(a). Above 1.9 GPa,  $T_c$  further decreases monotonically down to 10 K at 4.4 GPa but the decreasing rate becomes slower above 3.5 GPa as shown in Fig. 2(b).

Next, we show the  $M$ - $T$  curve for  $\text{FeSe}_{1-x}\text{Te}_x$  ( $x=0.7$ ) single crystals under various pressures measured using NaCl as PTM to generate uniaxial pressure along  $c$ -axis in Fig. 3(a) and along  $a$ -axis in Fig. 3(b). As seen in Figs 3(a) and 3(b), the  $M$ - $T$  curve at ambient pressure shows a sudden decrease below  $\sim 14$  K, indicating the onset of diamagnetic response. Also, the

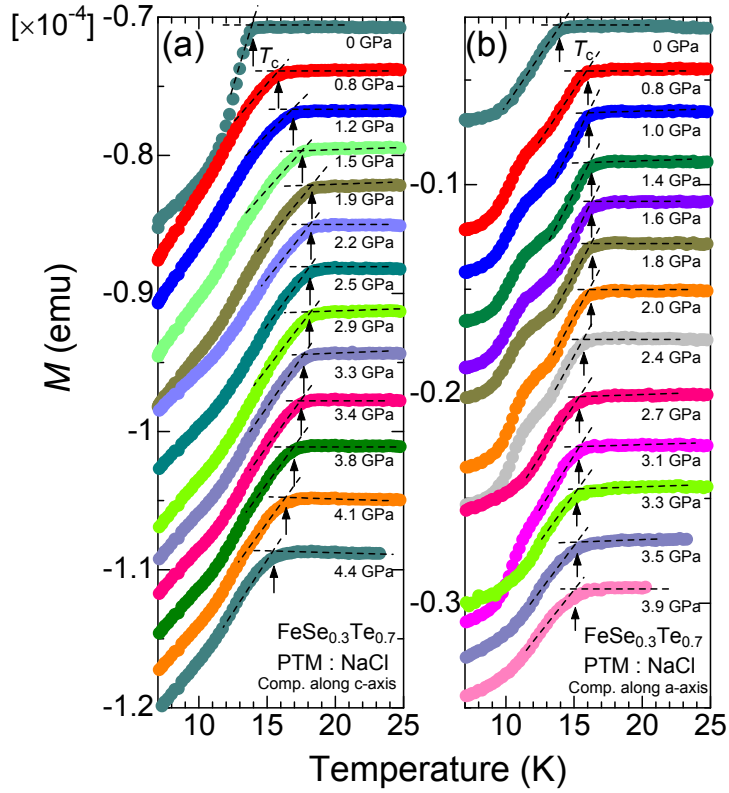


Figure 3: Temperature dependence of zero-field-cooled DC magnetization for  $\text{FeSe}_{1-x}\text{Te}_x$  ( $x=0.7$ ) measured in a magnetic field of 20 Oe under various pressures using NaCl as a pressure transmitting medium (PTM). Single crystal specimens were compressed along  $c$ -axis (a) and  $a$ -axis (b). The data are intentionally shifted along the longitudinal axis for clarity.

diamagnetic onset shifts toward higher temperature side by the application of uniaxial pressure along  $c$ - or  $a$ -axis, showing that  $T_c$  is enhanced to  $\sim 16$  K at  $P \sim 1$  GPa. The onset temperature shifts further to higher temperature side under uniaxial pressure along  $c$ -axis above 1 GPa in Fig. 3(a), whereas the onset is almost never changed under uniaxial pressure along  $a$ -axis above 1 GPa in Fig. 3(b). Figure 3(a) shows that the diamagnetic onset increases up to a maximum of  $\sim 18$  K by the application of uniaxial pressure along  $c$ -axis at 2.5 GPa but decreases gradually by the further pressure down to  $\sim 15$  K at  $P=4.4$  GPa.

In Fig. 4,  $T_c$  versus  $P$  data for the hydrostatic and nearly uniaxial compression (along  $a$ - and  $c$ -axis) are summarized. In the figure, the  $T_c$ - $P$  curves appear to be similar to each other especially below  $\sim 1$  GPa but depend on the hydrostaticity and direction of the compression above 1 GPa. Under hydrostatic pressure,  $T_c$  increases and makes a maximum of  $\sim 15$  K at  $\sim 1$  GPa, but decreases monotonically as increasing pressure down to  $\sim 10$  K at 3.5 GPa, above which  $T_c$  appears to be pressure independent. In contrast,  $T_c$  under uniaxial pressure along  $c$ -axis increases even above 1 GPa and makes a broad maximum of  $\sim 18$  K at  $\sim 2.5$  GPa, followed by the gradual decrease down to 15 K at 4.5 GPa. A striking feature is also seen in  $T_c$ - $P$  curve under uniaxial pressure along  $a$ -axis, where  $T_c$  is almost pressure independent above 1

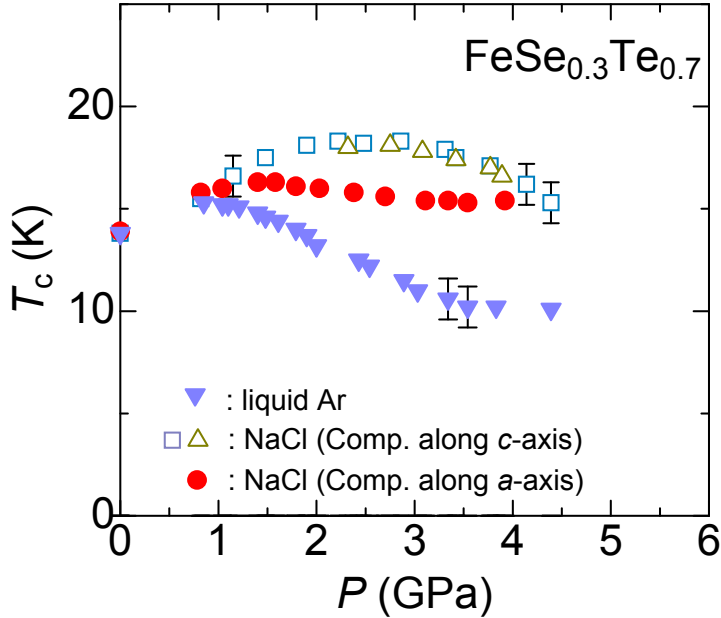


Figure 4: Plots of  $T_c$  versus pressure for  $\text{FeSe}_{1-x}\text{Te}_x$  ( $x=0.7$ ) using liquid Ar and NaCl as pressure transmitting media.

GPa. The  $T_c$ - $P$  curve under hydrostatic pressure indicates that isotropic compression of the crystal remarkably suppresses the superconductivity. The effect of the hydrostatic (isotropic) compression may be expected to be similar to the combined effect of uniaxial compression along  $a$ - and  $c$ -axes, but is actually not. This suggests that the effect of uniaxial compression along individual axis is not independent to each other, so that the simultaneous compression along  $a$ - and  $c$ -axes gives a quite different effect. On the other hand, the  $T_c$ - $P$  curve under uniaxial pressure indicates that the lattice compression along  $a$ -axis induces no remarkable change in the superconductivity, but that along  $c$ -axis enhances the superconductivity. We note that the uniaxial compression along  $c$ -axis in the present study enhances the superconductivity but also suppresses the superconductivity especially in high pressure region. This is probably because the compression along  $c$ -axis by using NaCl as PTM does not provide a pure uniaxial pressure, inducing also a pressure component along  $a$ -axis. If the specimen is compressed both along  $a$ - and  $c$ -axes, the superconductivity could be suppressed, as seen in the  $T_c$ - $P$  curve under hydrostatic pressure. The most challenging question is how much  $T_c$  will be enhanced by the compression purely along  $c$ -axis. Experiments using uniaxial pressure apparatus for magnetic measurements[19] and high quality single crystal specimens are highly desired in the future study.

## 4 Summary

In the present work, we have investigated the superconductivity under pressure in  $\text{FeSe}_{1-x}\text{Te}_x$  with  $x=0.7$  ( $T_c=14$  K) by the measurements of dc magnetization, applying external pressures

of different types, that is hydrostatic and nearly uniaxial pressure along  $c$ - and  $a$ -axis. It has been found that  $T_c$  decreases under hydrostatic pressure but increases (becomes constant) under uniaxial pressure along  $c$ -axis ( $a$ -axis) above 1 GPa, whereas  $T_c$  increases with similar slopes for  $0 \leq P \leq 1$  GPa. These results suggest that the isotropic compression remarkably suppresses the superconductivity but the anisotropic compression along  $c$ -axis ( $a$ -axis) enhances (never changes) the superconductivity.

## 5 Acknowledgment

This work is supported by the technical staff at Department of Materials Analysis, ICSR, Shimane University. The authors thank S. Nishigori and H. Katsube for experimental help.

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