Crystal orientation and thickness dependence of the superconducting transition temperature of tetragonal $FeSe_{1-x}$ thin films

Superconductivity was discovered in tetragonal $FeSe_{1-x}$, which has the simplest Fe planar structure, with a transition temperature T_C ~ 8 K [1]. It was noted that a low temperature structural distortion was observed in $FeSe_{1-x}$ superconductors [1] without accompanying a magnetic ordering, which is quite different from the Fe-pnictide superconductors. Wu *et al*. have reported the suppression of the superconductivity in Cu-substituted $Fe_{1-y}Cu_ySe_{1-x}$ samples, which have no low temperature structural distortion [2]. Cava *et al*. have also reported that a low temperature structural distortion is observed in the superconducting $Fe_{1+\delta}Se$ (δ~0.01) samples, but not in the non-superconducting $Fe_{1+\delta}Se$ (δ~0.03) samples [3]. This correlation suggested that the low temperature structural distortion might be essential for the occurrence of superconductivity. In this work [4], we further confirmed the importance of the low temperature distortion to the occurrence of superconductivity in $FeSe_{1-x}$ thin films, which exhibit strong orientation and film thickness dependence.

The superconducting tetragonal $FeSe_{1-x}$ thin films on (001) MgO substrates were grown using a pulsed laser deposition (PLD) technique. In this report, the films were grown at substrate temperatures of 320°C (referred to as LT-FeSe films) and 500°C (HT-FeSe films). The detailed structure analysis at low temperature was performed using 4-circle X-ray diffractometer with incident beam (12.4 keV) at beamlines BL13A in NSRRC and **BL12B2** in SPring-8.

Figure 1(a) shows the X-ray diffraction patterns of the LT-FeSe and HT-FeSe films. The thicknesses of both films are estimated to be ~140 nm. We observed in the LT-FeSe sample only (001) peaks of the tetragonal phase $FeSe_{1-x}$, suggesting a strong *c*-axis preferred orientation. However, the HT-FeSe film shows the (101) peaks to be dominant. It was surprising to find, as shown in Fig. 1(b), that the LT-FeSe film exhibits only a slightly resistive drop at 2 K without a complete superconducting transition. On the contrary, a clear superconducting transition takes place in the HT-FeSe films.

To understand this puzzling phenomenon on superconductivity suppression in the LT-FeSe thin film, a series of LT- and HT-FeSe films with different thicknesses were prepared. For the LT-FeSe films, as the film is made thicker, the onset T_C of film recovers more closely to the bulk result. On the other hand, the HT-FeSe film does not exhibit significant film thickness dependence. The superconducting transition temperatures for HT-FeSe films are close to the bulk sample value.

The detailed lattice constants of films determined from high resolution X-ray diffraction show that both the LT- and HT-FeSe samples have lattice constants close to the values of bulk, a=3.78 Å and c=5.54 Å at room temperature. Thus, the lattice mismatch is unlikely to be the cause for the strong thickness dependence of superconductivity in LT-FeSe films.

To better understand our observations, we carried out detailed low temperature structure characterizations

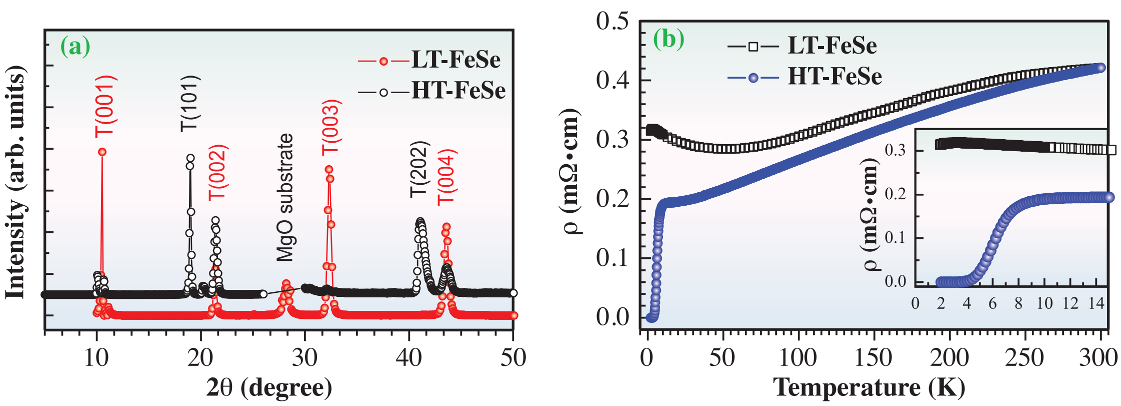

Fig. 1. **(a)** X-ray diffraction pattern of LT- and HT-$FeSe_{1-x}$ films. **(b)** ρ-T curves of LT- and HT-$FeSe_{1-x}$ films. Superconducting transition is observed only for HT-FeSe. The inset is a close-up of the ρ-T below 15 K.

at SPring-8, and the results are shown in Fig. 2. Figure 2(a) is the (220) peaks at different temperatures of the HT-FeSe film (thickness ~400 nm). The results clearly show the splitting of (220) peak confirming the presence of a structural distortion. On the other hand, the non-superconducting LT-FeSe film (~140 nm) showed no structural distortion at low temperature, as shown in Fig. 2(b). However, the distortion re-appears in thicker LT-FeSe film as evidenced by the gradual broadening of the (221) diffraction peak, and the maximum value is decreasing as the temperature decreases, as shown in Fig. 2(c). These observations clearly confirmed that the low temperature structural distortion is not affected in HT-FeSe films, but is suppressed in relatively thin LT-FeSe films. Our results further confirm the importance of the low temperature structural distortion to the origin of superconductivity in the FeSe system. Although the actual mechanism has yet to be identified, our results may provide an important clue to understand the origin of superconductivity in FeSe.

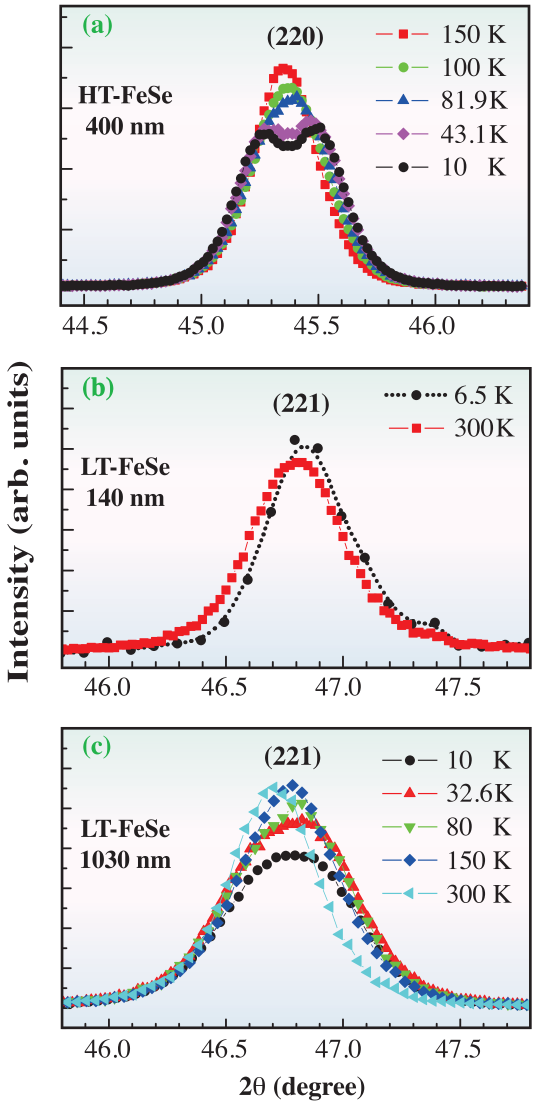

Fig. 2. **(a)** Low temperature X-ray diffraction (220) peaks of HT-FeSe film. The splitting of the Bragg peak indicates that structural distortion occurs at around 82 K. X-ray diffraction patterns of LT-FeSe films of **(b)** 140 and **(c)** 1030 nm thicknesses. The (221) Bragg peak does not change with temperature for the 140-nm-thick film, but broadens at low temperature for the 1030-nm-thick.

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