



# Significant enhancement of superconducting properties in the FeSe<sub>0.5</sub>Te<sub>0.5</sub> bulks by minor Sn addition



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## ABSTRACT

Sn addition improves bulk superconducting properties of FeSe<sub>0.5</sub>Te<sub>0.5</sub> with the increase of onset transition temperature and sharpens the transition width. The critical current density ( $J_c$ ) of Sn-added sample is about  $7 \times 10^3 \text{ A cm}^{-2}$  in self field and maintains a stable value of  $3 \times 10^2 \text{ A cm}^{-2}$  even beyond 60,000 Oe at 4.3 K, much higher than that of the Sn-free one. Phase compositions and morphology characterizations indicate that minor Sn addition facilitates the formation of the superconducting phase whose composition is closer to the ideal one, and suppresses the formation of Fe<sub>7</sub>Se<sub>8</sub> impurity and amorphous phase, which contribute to the improvement of  $J_c$ .

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## 1. Introduction

Iron-based superconducting compounds have revitalized the field of superconductors since they provide a new way to understand how the mechanism of the superconductivity differed from the cuprates [1–4]. The novel superconducting FeSe<sub>1-x</sub>Te<sub>x</sub> compounds, a simple system to study the iron-based superconductors, have attracted extensive attention [5–8]. The nontoxicity in composition and simplicity in structure favor investigations on the effects of chemical doping and high pressure experimentally and theoretically, contributing to a better understanding for the mechanism of the superconductivity and the promising applications of technology.

Chemical addition usually plays a significant role in enhancing the superconducting properties by facilitating the crystallization of the superconducting phase, reducing the amount of detrimental impurities, or introducing pinning centers. For example, an obvious improvement in transition temperature,  $T_c$ , can be achieved in MgB<sub>2</sub> since MgO impurity can be effectively attenuated by Cu addition [9] and a remarkable enhancement of  $J_c$  can be realized because of the modified grain dimension by Pb or Ag addition [10,11]. The textured grains induced by Sn addition [12] are also conducive to improving  $J_c$  in the Sr<sub>0.6</sub>K<sub>0.4</sub>Fe<sub>2</sub>As<sub>2</sub> tapes.

In present letter, we investigated the superconducting properties of nominal FeSe<sub>0.5</sub>Te<sub>0.5</sub> bulks with 5 wt% Sn addition in detail.

It was found that minor Sn addition facilitates the formation of the superconducting phase whose composition is closer to the ideal FeSe<sub>0.5</sub>Te<sub>0.5</sub>. Meanwhile, the content of detrimental impurity Fe<sub>7</sub>Se<sub>8</sub> and amorphous phase is decreased, and the homogeneity is improved. The electrical and magnetic results show a higher  $T_c^{\text{onset}}$  with much sharper superconducting transition as well as a remarkably enhanced  $J_c$  at high fields was obtained in the Sn-added sample.

## 2. Experimental

The nominal FeSe<sub>0.5</sub>Te<sub>0.5</sub> samples by Sn addition were prepared by solid state reaction route. The pressed cylinders of the stoichiometric powders of Fe (99.99%), Se (99.99%), Te (99.99%) were sintered at 550 °C for 12 h under the protection of pure argon gas. The Sn-free and 5 wt% Sn-added (99.9% purity) powders of the reacted mixture were pressed and treated at 600 °C for 5 h under the protection of pure argon gas. Detailed process can be found elsewhere [13]. The phase composition and the microstructure of the sintered samples were identified by the X-ray diffraction (XRD) and the high-resolution transmission electron microscopy (HR-TEM), respectively. The element mapping and EDX spectrums were detected by the scanning electron microscope (SEM) and the transmission electron microscopy (TEM). The resistivity and magnetization measurements were performed on a physical property measurement system (Quantum design, PPMS-14T).

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### 3. Result and discussion

The powder XRD patterns of the Sn-free and the Sn-added  $\text{FeSe}_{0.5}\text{Te}_{0.5}$  samples are shown in Fig. 1(a). The pattern of the main phase in both samples is well indexed to the superconducting tetragonal phase. Impurity phases, such as the hexagonal  $\text{Fe}_7\text{Se}_8$  phase and the oxide phase  $\text{Fe}_3\text{O}_4$ , which are generally observed in the sintered iron-based superconductors, are also detected. However, weaker  $\text{Fe}_7\text{Se}_8$  peaks are found in the Sn-added sample, accompanied by the appearance of a small amount of  $\text{SnSe}_{0.3}\text{Te}_{0.7}$  phase. As reported in the previous report [14], the existence of  $\text{Fe}_7\text{Se}_8$  exhibits a detrimental effect on the superconductivity performance. Since minor Sn suppresses the formation of  $\text{Fe}_7\text{Se}_8$ , it is expected a sharper superconducting transition for the Sn-added sample than for the Sn-free one.

As shown in Fig. 1(b), the shift to lower angles of (001) and (101) reflections in the Sn-added sample compared with the Sn-free sample indicates the increase of the lattice parameters for the tetragonal phase. According to the change of the angles, the lattice parameters were calculated, and the results are shown in the inset. The parameter varies from 3.7958 to 3.8007 Å for  $a$  and from 5.8994 to 5.9575 Å for  $c$  after 5% Sn addition. Since the added Sn exists as  $\text{SnSe}_{0.3}\text{Te}_{0.7}$ , it is speculated that the increased lattice parameters of the tetragonal phase are induced by the modulation of Se/Te ratios.

In order to verify the speculation above, average compositions of the tetragonal structure were detected by an EDX (Energy Dispersive X-ray Spectrometer). The final composition of the tetragonal phase in the Sn-free sample is 1.0:0.59:0.41, while 1.0:0.51:0.49 in the Sn-added one, which is closer to the nominal composition. This indicates that minor Sn addition facilitates Te substitution on Se-site of the tetragonal phase, and implies more favorable superconductivity performance.

The TEM photograph of the superconducting tetragonal phase is shown in Fig. 2(a). The upper inset shows part of a single tetragonal grain and the lower inset is the corresponding diffraction pattern with the electron beam parallel to [111]. As expected in the electron diffraction pattern, the (101) spots form an angle of  $106.5^\circ$  (the calculated value is  $106.8^\circ$ ) with the (011) spots, confirming the existence of single crystal of the superconducting phase.

Further observations of single tetragonal grains were performed by HR-TEM and the results are presented in Fig. 2(b) and (c). The measured lattice distances in both samples accord with the values of the tetragonal structure calculated from X-ray diffraction peaks. Amorphous phase was observed around the tetragonal grains in the Sn-free sample (Fig. 2(b)). In contrast, the grain boundary can be clearly observed in the Sn-added sample (Fig. 2(c)), which means Sn addition could suppress the formation of the

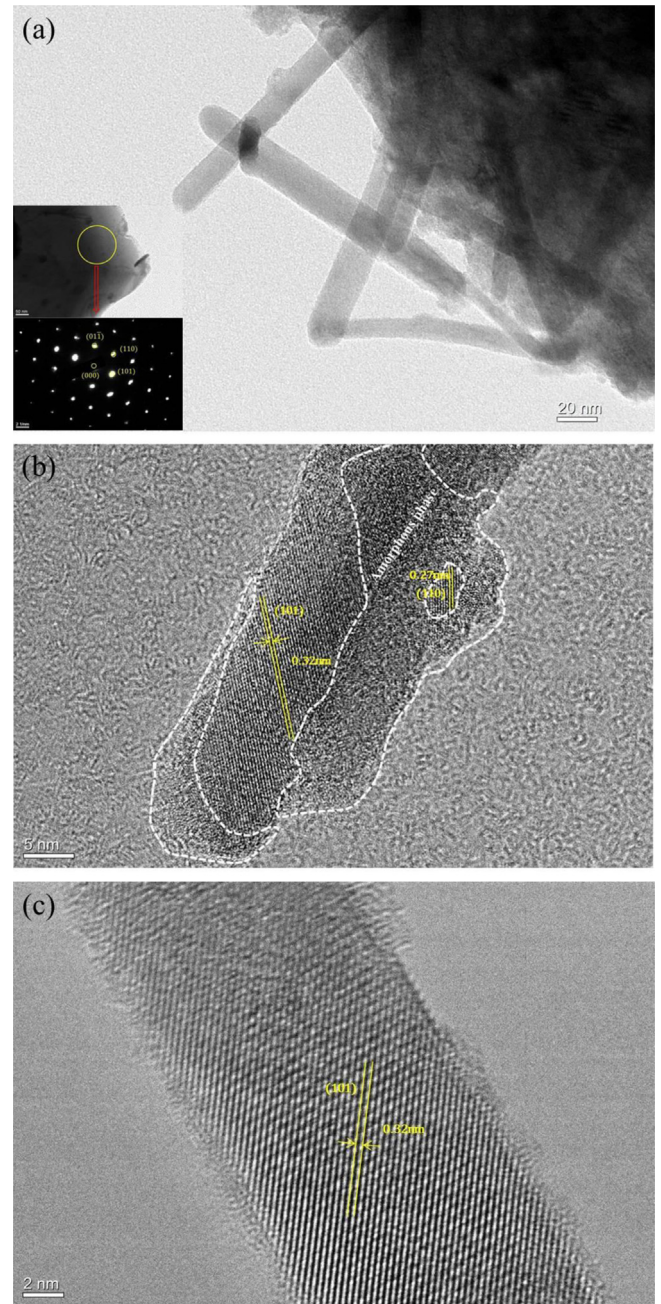


Fig. 2. (a) TEM image of the superconducting tetragonal phase in the Sn-added sample, and HR-TEM images of (b) the Sn-free and (c) the Sn-added samples.

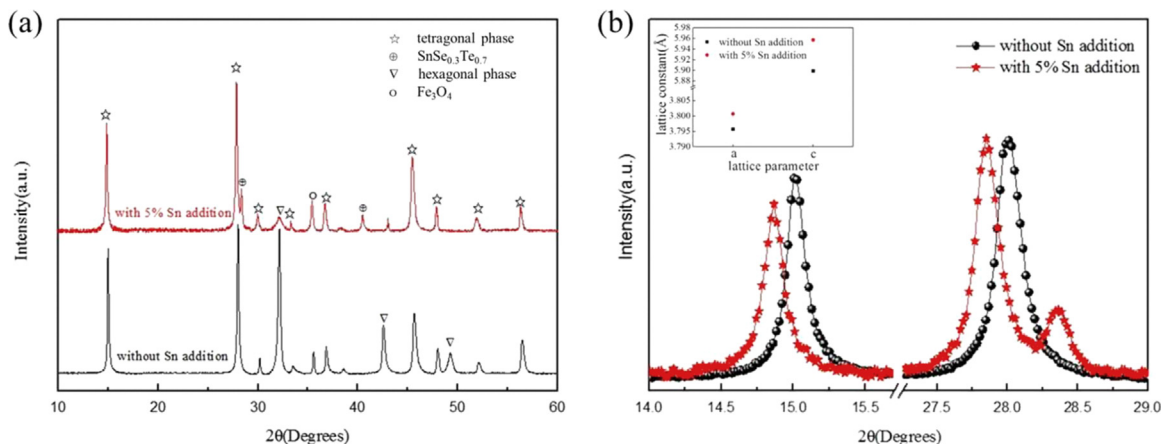


Fig. 1. (a) The powder XRD patterns of the Sn-free and Sn-added  $\text{FeSe}_{0.5}\text{Te}_{0.5}$  samples. (b) The comparisons of (001) and (101) reflections of the tetragonal phase in both samples. The inset of (b) shows the changes of the lattice parameters after Sn adding.

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