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The transport properties in antimony doped iron selenide $Fe(Se_{1-x}Sb_x)_{0.92}$ system

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ABSTRACT

The transport properties in the nominal composition $Fe(Se_{1-x}Sb_x)_{0.92}$ (x = 0, 0.05, 0.1, 0.2) system were studied. An abnormality was found on the resistivity curve which was ascribed to the structural transition. It was found that doping with Sb could change the transition temperature of structure transition T_s , while at the same time the change of superconducting transition temperature shows the same tendency. This may suggest that the structural transition and superconductivity has an internal relationship in the iron-based superconductors. Furthermore, two transport behaviors were found up and below structure transition like been reported in cuprates, organics, and 122-phase iron-based superconductors. This might be due to the magnetic order. At last, an insulator-metal transition was found at high temperature, suggesting a strong electron-phonon coupling in this system.

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1. Introduction

The discovery of superconductivity in iron-based LnOFeAs (1111-phase, Ln = rare earth metal) and MFe₂As₂ (122-phase, M = alkaline earth metal) has attracted great interests [1–5]. It also provides us a possibility of searching for novel superconductors with higher T_c and a way to get the point of high temperature superconducting mechanism. In high- T_c copper oxides, the parent compound has an AFM ground state. When it is doped with electrons or holes, the long-range AFM order is suppressed and then superconductivity appears. In iron-based superconductors, the phenomenon is more complex. It experiences a structure transition and an AFM order which is ascribed to the formation of spin density wave. Yet, the origin of superconductivity in iron-based compounds is still unclear. Many open questions are waiting to be solved especially the relationship between superconductivity, structural change and magnetic order.

Up to now, all the iron-based superconductors could be divided into three main categories, the 1111-phase, and 122-phase and iron selenide (11-phase), among them FeSe (11-phase) has the simplest structure. Also it is arsenic free. It is relatively easier to synthesize and study. Recently, the T_c of FeSe has been reported to reach as high as 36 K under pressure [6]. An alternative way to introduce pressure to change parameters and thereby affect superconductivity is by chemical substitutions.

In this work, we repot studies of the relationship between structural transition and superconductivity in Fe chalcogenide with a nominal formula $Fe(Se_{1-x}Sb_x)_{0.92}$ (x = 0, 0.05, 0.1, 0.2). We find that all $Fe(Se_{1-x}Sb_x)_{0.92}$ becomes superconducting with a structure tran-

sition around 100 K. Whereas the structure transition temperature changes with Sb doping, at the same time the change of superconducting transition temperature shows the same tendency. Two transport properties were observed which might be connected to the formation of magnetic order. Also, an insulator-metal transition was observed at high temperature region.

2. Experimental

We prepared $Fe(Se_{1-x}Sb_x)_{0.92}$ (x = 0, 0.05, 0.1, 0.2) by using a solid state reaction method. The $Fe(Se_{1-x}Sb_x)_{0.92}$ samples were synthesized as follows. First, the powders of Fe (99%), Se (99.95%), and Sb (99.99%) were sealed in an evacuated quartz tube with a nominal composition of $Fe(Se_{1-x}Sb_x)_{0.92}$ and were heated at 700 °C for 13 h. Following regrinding, the powders were pressed into pellets and heated in vacuo at 680 °C for an additional period of 40 h. Then all the samples were cooled with furnace. Finally, we obtained homogeneous black pellets. In this work, we denote the sample name as the composition, S1, S2, S3, S4 for FeSe_{0.92}, Fe(Se_{0.95}Sb_{0.05})_{0.92}, Fe(Se_{0.9}Sb_{0.1})_{0.92}, Fe(Se_{0.8}Sb_{0.2})_{0.92}, respectively. The samples were characterized by X-ray diffraction using Cu Ka radiation. Temperature dependence was measured from 380 K to 2 K using a four-probe method. The strain measurement was performed using a multifunction pole (MFP-05, metrical appurtenance of PPMS) and strain gauge with a data logger (TDS102, Tokyo Sokki Kenkyujo Co., Ltd.) as been described in Ref. [7].

3. Results and discussion

Fig. 1a–d shows the X-ray diffraction pattern for Fe(Se_{1-x}Sb_x)_{0.92}. Most of the peaks could be indexed using space group P4/nmm





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Fig. 1. (a-d) X-ray diffraction pattern of sample Fe(Se_{1-x}Sb_x)_{0.92}. (e-g) Change of lattice parameters with doping.

except some little impurity peaks. The impurity was found to be Fe_7Se_8 which is usually found in the preparation of 11-phase ironbased superconductors. Fig. 1e–g presents the parameter and volume vs. Sb doping. As can be seen, with the increase of Sb doping, the parameter a (b) decreases except at x = 0.1. The abnormality here will be discussed below.

Fig. 2 shows the temperature dependence of FeSe_{0.92} (S1), Fe(Se_{0.95}Sb_{0.05})_{0.92} (S2), Fe(Se_{0.9}Sb_{0.1})_{0.92} (S3), and Fe(Se_{0.8}Sb_{0.2})_{0.92} (S4). All samples show clearly superconducting transition at low temperatures. There are many ways to determine the critical temperature. For example, the onset temperature T_c^{onset} , zero resistivity temperature T_c^{cero} , midpoint temperature T_c^{onset} and so on. Here we define the critical temperature by using differentiation of $\rho(T)$ curve as shown in Fig. 3. The critical temperatures were found to be 7.6 K, 9.2 K, 8.3 K, and 8.85 K for S1, S2, S3, and S4, respectively. In order to further confirm the superconducting transition, we measured the temperature dependence of magnetization. The bot-



Fig. 2. Temperature dependence of resistivity for sample $Fe(Se_{1-x}Sb_x)_{0.92}$ (x = 0, 0.05, 0.1, 0.2). The inset shows an enlargement of the low temperature region.

tom inset of Fig. 4a shows the zero-field-cooling curve of temperature dependence of magnetization from 2 K to 100 K under 50 Oe for S3. From the M(T) curve, we can also confirm the superconducting transition.

Iron selenide has been reported to show a phase transition from tetragonal to orthorhombic around 90 K by different measurements [8,9] with an abnormality could be seen on the transport curve. Here we use the differentiation of $\rho(T)$ curve and strain measurement to determine its structure transition. As is shown in Fig. 4. by the derivation of the $\rho(T)$ curve of Fig. 4a, the phase transition could be determined in Fig. 4b which is 97 K. In iron-based superconductors when the structure changes from tetragonal to orthorhombic, the size of its volume would expand [10]. In Fig. 4c, with the decreasing of temperature the length of sample shrinks linearly, while at the temperature of phase transition it expands, then after the structure transition the shrinking behavior becomes linear again. The volume expanding temperature is consistent with T_s obtained from Fig. 4b. By using the method just described, the phase transition temperatures of S1, S2, S3, and S4 were determined to be 90 K, 100 K, 97 K, and 98 K, respectively.

As is known, FeSe_{1-x} experiences a structure transition between 70 and 100 K [8,9,11–13]. Yet in Fe-rich compositions, the structure transition is reported to disappear, and superconductivity is not found [12], suggesting that the structure transition is related to the occurrence of superconducting phase. Fig. 5 shows the *x* content dependence of critical temperature and phase transition. It is quite interesting that both T_c^{peak} and T_s have the same tendency with *x* changing which indicates that there might have some internal relationship between superconductivity and structure transition in iron selenide superconductors.

There have already been some reports on the linear resistivity in overdoped high- T_c cuprates, organic and 122-phase iron-based superconductors [14–18] which was thought to be driven by antiferromagnetic fluctuations. In Fe(Se_{1-x}Sb_x)_{0.92} the normal state resistivity shows two different transport behaviors up and below structure transition temperature (T_s). Under T_s the resistivity follows a strict linear behavior which could be well described by a fit of the form $\rho = \rho_0 + AT$ as can be seen in Fig. 6. However, above

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