Superconducting properties of FeSe wires and tapes prepared by a gas diffusion technique

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Abstract

Superconducting FeSe in the form of wires and tapes were successfully fabricated using a novel gas diffusion procedure. Structural analysis by means of x-ray diffraction shows that the main phase of tetragonal PbO-type FeSe was obtained by this synthesis method. The zero resistivity transition temperature of the FeSe was confirmed to be 9.3 K. A critical current density as high as 137 A cm$^{-2}$ (4 K, self-field) was observed. The results suggest that the diffusion procedure is promising in preparing high-quality FeSe wires and tapes.

(Some figures in this article are in colour only in the electronic version)

The discovery of superconductivity in the iron pnictides has triggered great interest in the past three years [1–7]. In addition to the high transition temperature, $T_c$, these Fe-based superconductors have been reported to have a very high upper critical field, $H_{c2}$, bringing hope in a wide array of future applications [8–10]. Up to now, several groups of iron-based superconductors have been discovered, such as LnOFeAs (1111 series) [1–3], BaFe$_2$As$_2$ (122 series) [5], LiFeAs (111 series) [11, 12] and FeSe (11 series) [6]. Among them, the 11 series has great advantages for applications due to the simple structure and containing no toxic arsenic. Very recently, by intercalating alkali metals into between the FeSe layers, superconductivity at around 30 K has been achieved [13–17]. Therefore, the FeSe-based materials deserve intensive studies for both fundamental physics and potential applications.

The common procedure for producing FeSe wires is the powder in tube (PIT) method [18, 19]. However, it is difficult to achieve high-density FeSe using the PIT process due to the package or the shrinkage of core materials. Hence, the transport $J_c$ of FeSe wire fabricated by this method is very low. An efficient technique to fabricate high-quality FeSe wires or tapes is still needed. As we know, the diffusion technique has been widely used for fabrication of superconducting tapes or wires such as Nb$_3$Sn [20], MgB$_2$ [21, 22], and so on. In this paper, a novel gas diffusion procedure was developed for preparing high-density FeSe superconductor.

The samples were prepared in the following way. Se powder and pure iron wires and tapes were used as raw materials. After cleaning carefully, the iron wires and tapes were cut into 4–6 cm pieces and sealed into an Fe tube with proper amounts of Se powder. Then these wires and tapes were reacted with Se vapor at temperatures varying between 400 and 800°C. The reaction time varied between 8 and 12 h. The final product was wires and tapes with an FeSe layer ranging from several microns to about 0.1 mm thick (figure 1), depending on the treatment process and the amount of Se powder. The FeSe layer can be easily separated from the iron wires and tapes. For further investigation, all the studied samples were taken from tape surfaces.

The phase identification and crystal structure investigation were carried out using x-ray diffraction (XRD). The temperature dependence of resistivity and transport critical current density $J_c$ were measured by a standard four-probe method using a physical property measurement system (PPMS). The microstructure was studied using scanning electron microscopy (SEM).

Figure 2 shows x-ray diffraction patterns for FeSe samples. Polycrystalline FeSe was obtained as a main phase.
Figure 1. Photograph of the final FeSe wires and tapes.

Figure 2. XRD patterns of the FeSe layer after peeling off the iron substrate. The impurity phases are marked by asterisks.

Figure 3. Temperature dependence of resistivity for an FeSe sample at zero-field up to 300 K. The inset shows the temperature dependence of dc magnetization for ZFC and FC processes at a magnetic field of $H = 20$ Oe.

Figure 4. SEM micrographs for the FeSe layers after peeling off the iron substrate. The cracks are indicated by arrows in (a).

With tetragonal PbO-type structure except for small amounts of impurity phases shown by asterisks. Hexagonal-NiAs-type FeSe was not observed in our samples.

Figure 3 displays the temperature dependence of electrical resistivity for FeSe samples with a measuring current of 1 mA. From this figure, a sharp drop in resistivity was observed below the onset temperature of about 15.1 K, and zero resistivity was attained below 9.3 K. This value is higher than the onset transition temperature for FeSe prepared by the solid state reaction method [6]. The normal state resistivity of our sample showed a broad bump at around 250 K and exhibited metallic behavior below 250 K. Similar behavior was also observed in the FeSe samples of [6]. The superconductivity of our sample was also confirmed by a DC magnetization measurement which is shown in the inset of figure 3. The relatively broad magnetic suggests that the inhomogeneity was still present in our sample.

In order to investigate the microstructure of our samples, an SEM microanalysis is employed in figure 4. It can be seen that the FeSe layer has a very dense structure with few cracks. The cracks probably originate from the difference in thermal expansion between the FeSe layer and iron. From the higher
magnification images we can clearly see a layered structure, very similar to what has been observed in other iron-based superconductors [10]. It is worth noting that textural structure was detected in some samples, as shown in figure 4(c).

The transport critical current density \( J_c \) as a function of temperature is presented in figure 5. A transport \( J_c \) as high as \( \sim 137 \, \text{A cm}^{-2} \) at 4 K and self-field has been observed in FeSe superconductors, much higher than the values for Fe(Se,Te) wires fabricated by the PIT method [18, 19], which have \( J_c \) values of only 12.4 cm\(^2\) and 64.1 A cm\(^{-2}\), respectively. The higher \( J_c \) values of the gas diffusion processed wires are presumably due to the higher FeSe layer density and the textural structure, as shown in figure 4. However, the \( J_c \) value of our sample is still lower than that obtained in the case of single crystal which generally attains \( 10^5 \, \text{A cm}^{-2} \) at 5 K [23]. Clearly, the impurity phases and the microcracks, which were observed in the XRD pattern and SEM image, are thought to be harmful to the transport capability. We expect that the transport \( J_c \) can be further increased by reducing the impurity phases, improving the texture or introducing flux pinning centers.

Figure 6(a) shows the resistive superconducting transitions for the FeSe sample under various magnetic fields. We tried to estimate the upper critical field (\( H_{c2} \)) and irreversibility field (\( H_{irr} \), using the 90% and 10% points on the resistive transition curves. The temperature dependences of \( H_{c2} \) and \( H_{irr} \) with magnetic fields up to 9 T for the FeSe samples were determined in this way and are shown in figure 6(b). It is clear that the curve of \( H_{c2} (T) \) is very steep with a slope of \( -dH_{c2}/dT \big|_{T_c} = 3.33 \, \text{K}^{-1} \), which significantly exceeds the Pauli limit of 1.84 K\(^{-1} \). The excellent \( H_{c2} (T) \) property indicates that this superconducting wire has encouraging applications in high fields.

We have prepared FeSe wires and tapes via the exposure of iron wires and tapes to Se vapor. Superconductivity with a zero resistivity transition temperature of 9.3 K was obtained. Transport \( J_c \) as high as \( \sim 137 \, \text{A cm}^{-2} \) at 4 K and self-field has been observed. The \( J_c \) value has much potential to be improved by perfect texture or optimization of the fabrication process. Our results clearly demonstrate that this synthesis technique is unique and simple, and hence will be able to be applied to the fabrication of the other iron-based superconducting wires, such as the FeAs 122 series or the recently discovered FeSe 122 series.

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