Global and local critical current density in superconducting SmFeAsO$_{1-x}$F$_x$ measured by two methods

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Abstract

The critical current densities of polycrystalline bulk SmFeAsO$_{1-x}$F$_x$ prepared by the powder-in-tube (PIT) method and by a conventional solid-state reaction were investigated using the remnant magnetic moment method and Campbell’s method. Two types of shielding current, corresponding to global and local critical current densities $J_c$ were observed using both measurement methods. The global and local $J_c$ were on the order of $10^7$ A/m$^2$ and $10^{10}$ A/m$^2$ at 5 K, respectively. The local $J_c$ decreased slightly with increasing magnetic field. The global $J_c$ was independent of the preparation method, while the local $J_c$ was larger for samples prepared by PIT than for those prepared by solid-state reaction.

Key words: FeAs based superconductor, critical current density, global

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and local $J_c$, remnant magnetization, Campbell’s method

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1. Introduction

The recent discovery of superconductivity in Fe-based compounds has attracted considerable attention, since the superconducting mechanism is different from that of Cu oxide superconductors, and Fe-based superconductors are expected to be useful in a wide array of future applications [1]. The critical temperature can reportedly be enhanced over 50 K by incorporating another rare earth element [2]-[4]. Although this value is still lower than the boiling point of liquid nitrogen (77.3 K), further discovery cannot be denied. In addition, it has been reported that the upper critical field may be higher than 100 T [5, 6]. Therefore, Fe-based superconductors may allow applications with higher operating temperatures.

We previously reported the preparation of SmFeAsO$_{1-x}$F$_x$. Polycrystalline bulk specimens were synthesized by conventional solid-state reaction [8] and by the powder-in-tube (PIT) method [9]. Recently a one-step synthesis was reported instead of the usual two-step synthesis [10].

Since the specimens were polycrystalline, it was expected that two types of shielding currents should be observed in the magnetization measurements [11]. They are known as local and global critical current densities. That is, connection between grains is too week and the difference of shielding current inside grains and through the grains are large as two to three order of magnitude. They are also called intra- and inter-grain shielding currents. However, the loop size of shielding current is not directly corresponding to the size of the grain for the case of very low quality polycrystalline specimen, since the shielding current may flow several grains or limited area of the grain. Therefore, in the present paper, the local and global critical current
densities are used instead of intra- and inter-grain critical current densities.

In our previous works [10], the local and global critical current densities are roughly estimated from the simple magnetization measurement. However, according to this method, the accurate measurement can not be expected since the measured magnetic moment is a type of average of the local and the global critical current densities [12]. Therefore, an accurate measurement for the local and global critical current densities is required.

In reference [11], the two kinds of shielding currents were estimated from the remnant magnetization as a function of maximum applied field, and it was explained that these currents corresponded to the local and global critical current densities. Although this measurement method is simple, the external magnetic field is restricted to zero, so it is impossible to obtain the magnetic field dependence of the critical current densities. In contrast, the two kinds of critical current density can be measured separately by the use of Campbell’s method [12, 13]. This method can much more effectively investigate the critical current densities in bulk specimens which have a complicated current path. Moreover, there are few reports on the characteristics of the critical current density of Fe-based superconductors.

In this paper, the two kinds of critical current density were measured using both the remnant magnetization method and Campbell’s method, with specimens prepared by two different preparation methods. The difference in values obtained by the two measurements is discussed.
2. Experimental

The sample specifications are listed in Table 1. Specimen #2 was prepared by the PIT method [9], and the other two specimens #3, #4 were prepared by conventional solid-state reaction with different nominal compositions of fluorine [8]. The details of their preparation are described in the references. The critical temperatures of the specimens, measured from the temperature dependence of magnetic moment in field cool and zero field cool, were about 50 K.

The structure of the specimens were examined by scanning electron microscopy (SEM). It was found that the typical grain size was about the same as was reported previously, i.e., in the range of 1–20 µm [8, 9]. There exists unreacted particles which size is smaller than the grain size. Although the connection between grains is almost insufficient, strong connected grains are also observed. That is, the size of loop shielding current is widely distributed. Therefore, the size of the loop is assumed as be 10 µm in the present study.

The remnant magnetic moment $m_R$ at zero magnetic field was measured after an excursion up to an external maximum magnetic field $H_a$. The shield current, which is identified as the critical current density $J_c$, is related to $H_a$. Assuming a superconducting slab of thickness $w$ and that the external magnetic field is applied in parallel to the wide surface, the remnant
magnetization $M_R$ is given as

$$M_R = \frac{H_a^2}{4H_p}; \quad H_a < H_p$$

$$= -\frac{H_a^2}{4H_p} + H_a - \frac{H_p}{2}; \quad H_p < H_a < 2H_p$$

$$= \frac{H_p}{2}; \quad H_a > 2H_p$$

where $H_p$ is the penetration depth which is equal to $J_c w / 2$ based on Bean’s model. Therefore, the derivative of Eq. (1) is given by

$$\frac{dM_R}{dH_a} = \frac{H_a}{2H_p}; \quad H_a < H_p$$

$$= -\frac{H_a}{2H_p} + 1; \quad H_p < H_a < 2H_p$$

$$= 0; \quad H_a > 2H_p.$$  

Hence, $dM_R/dH_a$ shows a peak at $H_a = H_p$, where $J_c$ can be estimated from the measured $H_p$. In the present study, remnant magnetic moment $m_R$ as a function of maximum magnetic field $H_a$ was measured using a SQUID magnetometer.

Campbell’s method was used to measure the critical current density of the specimens. The DC and superimposed AC magnetic fields were applied
to the specimen. The frequency of the AC magnetic field was 97 Hz, and the maximum AC magnetic field was 10 mT. The total magnetic flux $\Phi$ penetrating the specimen was calculated from the signals of pick-up and cancel coils as a function of the AC magnetic field amplitude $b_{ac}$. Then the penetration depth of the AC magnetic field $\lambda'$ was derived as

$$\lambda' = \frac{1}{2w \partial \Phi / \partial b_{ac}}. \quad (3)$$

The slope of the $\lambda'$-$b_{ac}$ plot gave $1/\mu_0 J_c$. The value of $\lambda'$ became saturated to a constant representing the center of the superconductor at a large $b_{ac}$ value. If there were two kinds of magnetic moments in the specimen, two slopes would be observed and two different critical current densities could be evaluated. Temperature of specimen was controlled by cryocooler above 18 K.

3. Results and Discussion

Fig. 1(a) shows the external applied magnetic field dependence of the derivative of the magnetic moment of specimen #2 at various temperatures. Two peaks were clearly observed. The same behavior was observed for specimen #3, which was prepared by solid-state reaction unlike the PIT preparation of #2. The first peak at smaller $H_a$ is related to the global critical current density, since the peak rapidly shifted to a lower magnetic field with increasing temperature, and the estimated value of $J_c$ was much smaller than the local critical current density. It was reported that the global critical current density measured by four probe method reached zero at temperatures higher than 20 K \[9\]. In contrast, the second peak at larger $H_a$ is related to
the local critical current density. It was found that the temperature dependence of the local critical current density was smaller than that of the global critical current density.

On the contrary, only one peak was found for specimen #4, as shown in Fig. 1(b). This peak corresponds to the local critical current density. The global critical current density of specimen #4 was too small to be measured under the present conditions.

The global and local $J_c$ evaluated from the peak positions of the derivatives of $m_R$ are shown in Fig. 2. Since only one peak was found for specimen #4, the global $J_c$ was omitted. The value of the global $J_c$ was on the order of $10^7$ A/m$^2$ at 5 K and could not be estimated for temperatures above 20 K, since the peak in the derivative of $m_R$ was too small. This value agrees with that reported by Yamamoto et al. [11]. The temperature dependence of global $J_c$ in specimens #2 and #3 were the same. Therefore, the global $J_c$ was too small and was significantly affected by voids and weak links between grains in the polycrystalline bulk specimens, both of which are known to exist in Cu oxide superconductors with poor properties. It is well-known that the temperature and magnetic field dependences of $J_c$ affected by weak links are large.

In contrast, the local $J_c$ at 5 K was over $10^{10}$ A/m$^2$, 1000 times larger than the global $J_c$, which was consistent with the results estimated in ref. [11]. It was found that the local $J_c$ was quite dependent upon the method of synthesis, as shown in Fig. 2(b). Moreover, the temperature dependence of local $J_c$ was also different. That is, the local $J_c$ of specimen #2 prepared by PIT was higher than specimens #3 and #4 prepared by conventional solid-
state reaction. Therefore, it is reasonable to expect that the characteristics of the critical current density could be improved by further developing the preparation method.

Fig. 3 shows an example of $\lambda'$ as a function of $b_{ac}$ measured using Campbell’s method. There were two slopes, corresponding to very-low and high $b_{ac}$. According to Campbell’s method, the first and second slopes are related to the global and local $J_c$, respectively. It was found that the global $J_c$ was only observed at zero DC magnetic field by Campbell’s method. That is, the global $J_c$ was very small, and the magnetic field dependence of $J_c$ was extremely poor, as is frequently observed as weak link property in Cu oxide superconductors [14]. This result agrees with that obtained by measurement of the remnant magnetic moment.

The temperature dependence of the local $J_c$ for specimen #2 at zero DC magnetic field estimated using Campbell’s method is also shown in Fig. 2(b). Although this value of local $J_c$ was quantitatively different from that obtained by the remnant magnetic moment method, the order of magnitude was the same. The reason for the quantitative difference is mainly caused by the difference of measurement methods. For example, although the change of the superconducting volume fraction does not affect to $J_c$ by the remnant magnetic moment method, it causes large affect to Campbell’s method. In addition, the local fluctuation of $J_c$ can be measured by Campbell’s method, while the measured $J_c$ is an average value by the remnant magnetic moment method.

Fig. 4 shows the magnetic field dependence of local $J_c$ at various temperatures, as measured using Campbell’s method. The values of local $J_c$ ranged
around $10^9 \text{ A/m}^2$, consistent with the prediction based on the remnant magnetic moment method. The local $J_c$ slightly decreased with increasing magnetic field and temperature, except in the temperature region near $T_c$, where a peak effect appeared in the magnetic field dependence of local $J_c$.

To investigate the value of $J_c$ over a wide range of magnetic fields, $J_c$ was estimated from the hysteresis of magnetic moment $\Delta m$ at various temperatures, measured using a SQUID magnetometer as shown in Fig. 5. This $J_c$ is a type of average of the local and the global $J_c$, and is different from both local and global $J_c$ [12]. The peak effect was widely observed at various temperatures. The first peak field was at about 0.1 T and was consistent with the result from Campbell’s method, as shown in Fig. 4. It is known that the peak effect is caused by the transition of the vortex lattice in Cu oxide superconductors [15]-[18]. However, the reason for the first peak effect in present case is unknown.

On the other hand, the second peak field was observed at magnetic fields of over 1 T. This seems unrealistic, since $J_c$ was observed at 50 K, which is higher than the critical temperature of specimen #3. The results of second peak effect and observed $J_c$ at 50 K could be ascribed to the possible existence of unreacted iron in the specimens. Since iron is ferromagnetic, the hysteresis of magnetic moment due to the ferromagnetism would remain, even at temperatures higher than $T_c$, and would disappear at magnetic fields larger than 1 T. This was confirmed by the temperature dependence of the magnetic moment $m$ in field cool, in which $m$ showed a positive value, even in the superconducting state. These different effects of residual ferromagnetic iron are the origin of the observed difference in local $J_c$ between the two
measurement methods, as shown in Fig. 2(b). Therefore, it is necessary to accurately estimate the inference of ferromagnetic property in the specimens.

4. Conclusion

In this work, two preparation methods were used to prepare specimens of polycrystalline bulk SmFeAsO$_{1-x}$F$_x$. The global and local shielding current densities, which correspond to the global and local critical current density, respectively, were measured using the remnant magnetic moment and Campbell’s methods. The global critical current density was very low, on the order of $10^7$ A/m$^2$ at 5 K, and this value was independent of the preparation methods. This is likely due to the effects of voids and weak links between grains in the polycrystalline bulk specimens. In contrast, the local $J_c$ was about 1000 times larger than the global $J_c$, and the value and temperature dependence of $J_c$ was largely dependent upon the preparation methods. The estimated $J_c$ from the two measurements were in agreement. Our results show that remnant unreacted iron in the specimens significantly affected the results of the $J_c$ measurements.

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| specimen No. | specimen     | synthesis                        | critical temperature [K] |
|-------------|--------------|----------------------------------|-------------------------|
| #2          | SmFeAsO$_{0.7}$F$_{0.3}$ | PIT (powder in tube)          | 50.4                    |
| #3          | SmFeAsO$_{0.7}$F$_{0.3}$ | solid state reaction         | 48.5                    |
| #4          | SmFeAsO$_{0.6}$F$_{0.4}$ | solid state reaction         | 51.1                    |
Figure captions

Figure 1 Derivative of remnant magnetic moment as a function of maximum external applied magnetic field at various temperatures in (a) specimen #2 and (b) specimen #4.

Figure 2 (a) Global and (b) local critical current densities as a function of temperature at zero magnetic field, estimated from remnant magnetic moment. Filled squares denote the results obtained using Campbell’s method.

Figure 3 Example of penetration depth of the AC magnetic field as a function of AC magnetic field by Campbell’s method. The inset shows an enlarged plot of the small AC magnetic field region.

Figure 4 Magnetic field dependence of local critical current density of specimen #2 at various temperatures obtained using Campbell’s method.

Figure 5 Magnetic field dependence of the critical current density, directly evaluated from the hysteresis of the magnetic moment of specimen #3 at various temperatures.
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Fig. 2: E.S. Otabe et al.
Fig. 3: E.S. Otabe et al.
Fig. 4: E.S. Otabe *et al.*
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