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Magnetic Field-Temperature Phase Diagram of Fine $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ Ceramics from Linear and Non-linear Resistivities

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We performed linear and non-linear resistivity measurements for fine $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ ceramics under magnetic field. Successive superconducting transition was observed in the linear resistivity and the lower temperature part of the resistivity curve became convex upward at high fields. A non-linear resistivity peak was observed for this lower temperature region and its offset temperature T_{offset} and the onset temperature T_{onset} coincided with the temperature of zero resistivity and inflexion point in linear resistivity, respectively. At lower field, T_{onset} was less sensitive to the field than T_{offset} , which implies the existence of the peculiar inter-grain ordering phase at finite fields.

KEYWORDS: fine ceramics superconductor, inter-grain ordering, non-linear resistivity, field-temperature phase diagram

1. Introduction

Fine ceramics superconductor has been attracted much attention since it shows unique features such as a successive superconducting transition derived from submicron grains and the Josephson-coupled networks [1]. This transition is explained by the intra-grain superconducting ordering at higher temperature region and the inter-grain ordering at lower temperature region. In the previous work, we investigated the fine ceramics materials composed of stoichiometric $\text{YBa}_2\text{Cu}_4\text{O}_8$ (Y124) submicron grains and reported the oxidation and reduction effects on the superconducting transition [2]. These effects were mainly seen in the inter-grain ordering and the change of the transition temperatures varied in response to the grain size, which suggests that the oxygen vacancy is induced on the grain surface. For further research, we performed a non-linear resistivity measurement. This measurement is suitable for detecting a subtle change at around the phase transition. In an ideal situation, current and voltage obey Ohm's law, however, they sometimes deviate from the proportional relationship because of the emergence of the

higher-order terms. The non-linear resistivity corresponds to these terms. In the fine Y124 ceramics, we observed a non-linear resistivity peak at around the inter-grain ordering transition and analyzed the oxidation and reduction effects on the peak width [2].

In this study, we performed the simultaneous measurements of the linear and non-linear resistivity for fine $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (Y123) ceramics under magnetic field to clarify the relation between the non-linear resistivity peak and the inter-grain ordering transition.

2. Experimental

Fine Y123 ceramics were prepared by the coprecipitation method which can make homogeneous grains of a few micron size. The grain size was estimated by the scanning electron microscope (SEM) images as shown in Fig. 1. The electrical resistivity measurements were performed by 4-probe method applying AC current of 23.00 Hz under magnetic field up to 12.98 kOe. The amplitude of the AC current was adjusted to set the current density to 9.5 mA/mm^2 . The linear and non-linear resistivities were defined as the coefficients of the first and the third power term of the voltage response, respectively.

3. Results and Discussion

Figure 2 (a) shows the temperature dependence of the linear resistivity in the bulk Y123 ceramics synthesized by the solid reaction method and the fine Y123 ceramics. T_C^{onset} denotes the onset of the superconducting transition temperature observed in the linear resistivity. Both ceramics show the superconducting drop at about 92 K. The resistivity curve in the fine ceramics branches away from that in the bulk ceramics at T_0 and the temperature of zero resistivity T_C^{zero} shifts from 85.1 K to 66.8 K. The

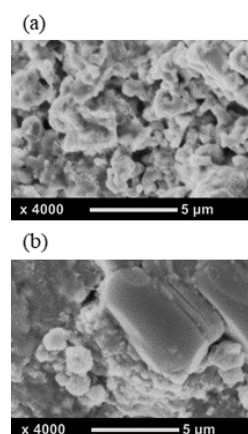


Fig. 1. SEM images for (a) the fine Y123 ceramics prepared by the coprecipitation method and (b) the bulk Y123 ceramics prepared by the solid reaction method. The grain size was estimated to be about $3 \mu\text{m}$ for the fine ceramics.

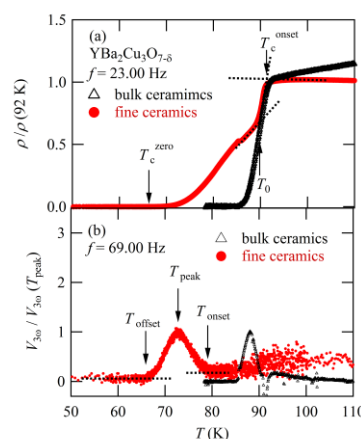


Fig. 2. Temperature dependence of (a) the linear resistivity and (b) the non-linear resistivity in the bulk and fine Y123 ceramics. The values of the linear and the non-linear resistivities are normalized to the value at T_C^{onset} and T_{peak} , respectively.

simultaneously observed non-linear resistivity is shown in Fig. 2 (b). Although the superconducting peak was observed for bulk and fine ceramics, the transition temperatures were different. Comparing Fig. 2 (a) with (b), the offset temperature of the non-linear resistivity peak T_{offset} coincides with T_C^{zero} in the linear resistivity, which suggests the non-linear resistivity peak reflects mainly the inter-grain superconducting transition.

Figure 3 (a) shows the temperature dependence of the linear resistivity under magnetic field. T_C^{onset} and the superconducting drop above T_0 show little change up to 12.98 kOe, whereas T_C^{zero} is suppressed by the field more than 40 K and the lower temperature part of the resistivity curve becomes convex upward at high fields. This change is attributed to the difference in the critical field of the inter-grain and intra-grain superconducting orderings and the two steps of the superconducting drop are expected to be clearer at higher fields. The simultaneously observed non-linear resistivity is shown in Fig. 3 (b). In contrast with the linear resistivity, the whole non-linear resistivity peak shifts toward lower temperature and the higher temperature part of the peak broadens with increasing the field. The peak could not be observed at high fields above 9.87 kOe because of the increase in the background noise.

The step-like linear resistivity curve in Fig. 3 (a) suggests that the onset temperature of the inter-grain ordering should be determined not from T_0 but from the inflexion temperature T^* in the convex curve. Therefore, we calculated the derivative of the linear resistivity. Figure 4 (a) shows the temperature dependence of $d\rho/dT$ observed at 976 Oe. The peak temperature T^* was estimated by fitting to normal distribution. As shown in Fig. 4 (b), T^* coincides with the onset temperature of the non-linear resistivity peak T_{onset} . This coincidence was observed at other field magnitudes too, thus both T^* and T_{onset} can be regarded as the onset temperature of the inter-grain ordering. This finding enables us to make the field-temperature phase diagram for the inter-grain superconducting ordering. Figure 5 shows the field dependence of T_{offset} , T_{peak} , T_{onset} , and T_C^{zero} at (a) high field up to 3.92 kOe and (b) low field up to 482 Oe. The transition lines for these transition temperatures are almost parallel at high field, however, T_{onset} in

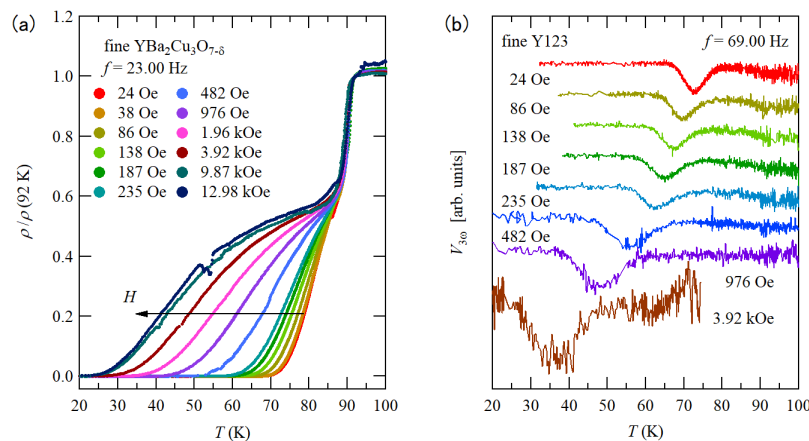


Fig. 3. (a) Temperature dependence of the linear resistivity under magnetic field up to 12.98 kOe. The resistivity curve becomes convex upward at high fields. (b) Temperature dependence of the non-linear resistivity under magnetic field up to 3.92 kOe. The higher temperature part of the peak broadens at high fields.

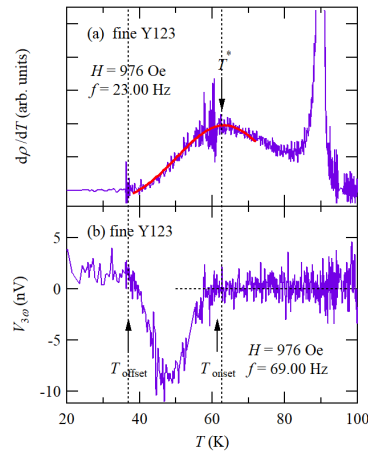


Fig. 4. (a) Temperature dependence of $d\rho/dt$ at $H = 976$ Oe. The peak temperature T^* was estimated by fitting to normal distribution. (b) Temperature dependence of the non-linear resistivity at $H = 976$ Oe.

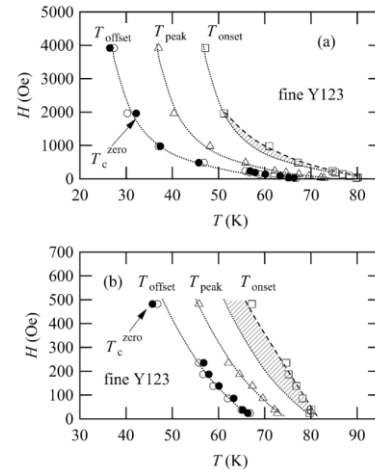


Fig. 5. Field-temperature phase diagrams for fine Y123 ceramics up to (a) 3.92 kOe and (b) 235 Oe. The dotted lines are guide for eyes drawn in parallel with T_{peak} curve. T_{onset} is less sensitive to the field compared with T_{offset} , T_{peak} , and $T_{\text{C}}^{\text{zero}}$ at low field. As a result, a shaded area appears below 2000 Oe.

Fig. 5 (b) is less sensitive to the field compared with T_{offset} , T_{peak} , and $T_{\text{C}}^{\text{zero}}$. This behavior is reminiscent of the field-temperature phase diagram reported for fine Y124 ceramics [3]. Deguchi *et al.* determined the transition temperatures $T_{\text{C}2}$ and $T_{\text{C}3}$ from the AC susceptibility and the linear resistivity, respectively. They estimated the power law exponent α from the field dependence of $|T_{\text{C}3}(0) - T_{\text{C}3}(H)|^\alpha$ and reported that the estimated value of $\alpha = 2.3$ is consistent with the spin-glass transition line predicted by Kawamura *et al.* [4] On the other hand, $T_{\text{C}2}$ was larger than $T_{\text{C}3}$ in an applied field and the transition line of $T_{\text{C}2}(H)$ at around 100 Oe corresponded with the Gabay-Toulouse (GT) line of the mean-field model, which suggests that the chirality is decoupled from the phase of superconducting order and a chiral-glass ordering takes place at $T_{\text{C}2}$.

In our study, the downward curved convex line for T_{peak} (T_{offset} , $T_{\text{C}}^{\text{zero}}$) and the upward curved one for T_{onset} at low field were similar to the transition lines of $T_{\text{C}3}$ and $T_{\text{C}2}$ in fine Y124 [3]. As shown in Fig. 5, writing the dotted line parallel to the transition line of T_{peak} , a shaded area like the chiral-glass phase appears below 2000 Oe. This implies the existence of the characteristic phase concerning the inter-grain ordering. However, the estimated values of α for T_{peak} and T_{onset} below 482 Oe were 1.2 and 0.85, respectively, which were not consistent with the spin-glass transition line nor the GT line. To clarify the mechanism of the inter-grain ordering, further magnetic measurements are desired.

4. Conclusion

We investigated the temperature dependence of the linear and non-linear resistivity under magnetic field. The linear resistivity curve becomes convex upward at high fields. The inflexion temperature T^* was determined from the derivative of the linear resistivity and it coincides with the onset temperature T^{onset} of the non-linear resistivity peak. The

field dependence of T^{onset} was different from that of the zero resistivity temperature, which implies the existence of the characteristic phase concerning the inter-grain ordering.

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