AC susceptibility measurements on ceramic YBa$_2$Cu$_3$O$_{7-y}$ superconductor in the GPa region

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Abstract. Ceramic high-$T_c$ superconductor has intra-grain superconducting transition at $T_{C1}$ and the inter-grain one at $T_{C2}$. We investigated the effects of pressure on the ceramic superconductor YBa$_2$Cu$_3$O$_{7-y}$ in the pressure region up to 5.3 GPa, through the AC susceptibility measurements using a miniature diamond anvil cell (mDAC). The $T_{C1}$ increases against the initial pressure, whereas it turns to decrease at around 2.5 GPa. The $T_{C2}$ also exhibits similar pressure dependence. The up-and-down change of $T_{C1}$ has been already observed in the previous single crystal experiment, whereas that of $T_{C2}$ was observed through the present study for the first time. From the viewpoint of measurement technique, we also point out some problems on the high pressure AC susceptibility measurements using a SQUID magnetometer.

1. Introduction

A ceramic high-$T_c$ superconductor exhibits the intra-grain and inter-grain superconducting transitions [1, 2]. The ceramic high-$T_c$ superconductors can be viewed as a weakly coupled random Josephson network. In the cooling process, the first superconducting order occurs inside each grain at $T_{C1}$ and the second one does between the grains at $T_{C2}$. The $T_{C2}$ value depends on the strength of Josephson coupling between grains, and superconducting phenomena at $T_{C2}$ have been much attention from the viewpoint of the chiral glass mechanism [3]. The $T_{C2}$ is influenced by the conditions of sample preparation such as temperature and pelletizing pressure [4]. Furthermore, it is known that “hydrostatic pressure” can be a physical parameter to control the strength of the Josephson couplings: In YBa$_2$Cu$_4$O$_{8-y}$, Deguchi et al. observed the enhancements of $T_{C1}$ and $T_{C2}$ as $dT_{C1}/dP = 5.5$ K/GPa and $dT_{C2}/dP = 7.0$ K/GPa in the pressure ($P$) region up to 0.8 GPa via the AC susceptibility measurement [5]. As for YBa$_2$Cu$_4$O$_{7-y}$, Koyama et al. reported $dT_{C1}/dP = 1.9$ K/GPa and $dT_{C2}/dP = 8.0$ K/GPa via similar measurements ($P \leq 0.6$ GPa) [6]. It is interesting that the value of $dT_{C2}/dP$ is independent of the initial value at ambient pressure and the empirical formula [6]. These pressure dependences about 0.8 GPa have not been investigated. Herein, we investigate the pressure dependences of both $T_{C1}$ and $T_{C2}$ of YBa$_2$Cu$_4$O$_{7-y}$ ceramics in the GPa region up to 5.3 GPa.

As for a convenient high pressure apparatus, Mito et al. developed a miniature diamond anvil cell (mDAC), which can be inserted into a SQUID magnetometer (Quantum Design, MPMS) [7, 8]. Most
of the parts are made of CuBe, and the overall cross-sectional view of the latest model is shown in Fig. 1 (a). The previous experiments have been performed in the style of the DC measurement [7-10]. Here, both $T_{C2}$ and $T_{C1}$ are detected by the AC susceptibility measurement. We will also describe some problems and their solutions on the AC susceptibility measurements using mDAC in MPMS.

![Overall cross-sectional view of the miniature diamond anvil cell](image)

**Figure 1. a:** Overall cross-sectional view of the miniature diamond anvil cell for a SQUID magnetometer. The main parts are (a) grand nut, (b) half-sphere seat for the upper diamond, (c) cylinder, (d) diamonds, (e) anvil plate for the lower diamond, (f) piston, (g) nut for applying pressure, (h) screw for tilt adjustments, (i) gasket, (j) support for the gasket, (k) screw for X-Y adjustments and (l) piston guide screw. **b:** Temperature dependence of in-phase and out-of-phase components of the first harmonic susceptibility at $H_{AC} = 0.5$ Oe and $f = 10$ Hz.

2. Experimental

In order to detect the magnetic response as much as possible in the present experiment, we used diamond anvils with the top diameter of 1.0 mm, resulting in restricting the pressure range within at most 6 GPa. The sample hole of 0.5 mm diameter was drilled in the CuBe gasket with the thickness of 0.2 mm. Small pieces of YBa$_2$Cu$_3$O$_{7-y}$ and a Apiezon-J grease as the pressure transmitting medium with a few crystals of ruby were placed into the sample hole. The pressure was estimated at room temperature according to ruby fluorescence technique [11]. The AC susceptibility was measured by the use of MPMS with the AC option. The inter-grain superconducting transition becomes unstable under the AC filed with large amplitude and high frequency. In previous studies of ref. 5 and 6, the AC field amplitude ($H_{AC}$) was 0.05 Oe and the frequency ($f$) was 10 Hz. In the present experiment, the value of $H_{AC}$ was increased to 0.5 Oe to obtain the SQUID response with bearable S/N ratio, resulting in reducing the slightly from the intrinsic value of $T_{C2}$. At $H_{AC} = 0.5$ Oe and $f = 10$ Hz, $T_{C1}$ and $T_{C2}$ at ambient pressure are 91 K and 30 K, respectively. Three runs of measurements were performed: Each maximum pressure was 3.5 GPa, 5.3 GPa and 4.6 GPa.

When the pick-up coil is located in the outside of the metal-made cell, the effect of the eddy current must be considered under the AC field. The magnitude of the phase shift depends on the AC frequency, and the calibration work must be performed prior to the spectrum analysis. We have already constructed the database on the adequate phase calibration via many AC susceptibility measurements about high pressure. The magnetic signal of the target sample with the mass of at most 100μg is much smaller than that of mDAC.

Figure 1(b) shows the in-phase ($\chi_1$) and out-of-phase ($\chi_2$) components of mDAC in the MPMS system at $H_{AC} = 0.5$ Oe and $f = 10$ Hz. The response of $\chi_1$ originates from the magnetically vacuum space (near diamond anvils) in the paramagnetic background, and the temperature dependence reflects the paramagnetism of CuBe. It is well-known that the electric conductivity on Cu exhibits residual
resistance below 30 K, whereas the electrical resistance increases with the linear fashion against temperature above 30 K. The $\chi_{1\omega}'$ just reflects the temperature dependence of the eddy current, which strongly depends on the electric conductivity of CuBe. The magnitude of $\chi_{1\omega}'$ is one order larger than that of $\chi_{1\omega}''$. However, the magnetic signal of mDAC fortunately slips out with the difference of about one-fourth cycle again the applied AC field. By using the imaginary Fourier transformation, we can distinguish the $\chi_{1\omega}'$ of sample signal and mDAC from the eddy current contribution of mDAC. Finally, the $\chi_{1\omega}'$ of the target sample can be derived after the correction of the paramagnetic behavior of mDAC.

3. Results and Discussion

Figures 2 and 3 show temperature dependences of $\chi_{1\omega}'$ in the first and the third runs, respectively. We observed that the pressure-induced enhancements of $T_{C1}$ and $T_{C2}$ saturate at around 2-3 GPa via three runs.

The pressure dependences of $T_{C1}$ and $T_{C2}$ are summarized in Fig. 4. For $P \leq 1.1$ GPa, we obtained $dT_{C1}/dP = 0.7$ K/GPa and $dT_{C2}/dP = 3.6$ K/GPa, which are smaller than the reference value by Koyama et al. [6]. The difference of $H_{AC}$ and the pressure quality may bring about the above-mentioned deviation. In the previous single crystal experiment, Klotz et al. have reported that the increase of $T_C$ saturates at around 4 GPa [12]. The present change of $T_{C1}$ was easily supposed.

![Figure 2](image1.png)

**Figure 2.** Temperature dependence of the AC susceptibility $\chi_{1\omega}'$ of the ceramic superconductor YBa$_2$Cu$_3$O$_{7-\delta}$ in the first run (a). The data at around $T_{C1}$ are enlarged in the right figure (b).

![Figure 3](image2.png)

**Figure 3.** Temperature dependence of the AC susceptibility $\chi_{1\omega}'$ of the ceramic superconductor YBa$_2$Cu$_3$O$_{7-\delta}$ in the third run (a). The data at around $T_{C1}$ are enlarged in the right figure (b).
Figure 4. Pressure dependences of $T_{C1}$ and $T_{C2}$ of the ceramic superconductor YBa$_2$Cu$_3$O$_{7-y}$.

The effects of pressure on the intra-grain superconductivity originate in the intrinsic feature of YBCO superconductivity, and maybe depend on the charge carrier and/or the strength of antiferromagnetic network on CuO$_2$ plane. On the other hand, the inter-grain superconductivity depends on the inter-grain Josephson coupling as well as the stability of intra-grain superconductivity. The up-and-down change of $T_{C2}$ at around 2.7 GPa probably originate in the instability of intra-grain YBCO superconductivity, since $T_{C1}$ and $T_{C2}$ show quite similar behaviors. However the enhancement of $T_{C2}$ under pressure was larger than that of $T_{C1}$. We can recognize that the strength of inter-grain Josephson coupling was also enlarged by artificial hydrostatic shrinkage.

In summary, we investigated the effects of pressure on the ceramic superconductor YBa$_2$Cu$_3$O$_{7-y}$ in the pressure region up to 5.3 GPa by the AC susceptibility measurements using mDAC in MPMS. The up-and-down changes of $T_{C1}$ and $T_{C2}$ were observed. These effects can be understood with the pressure dependences of the intrinsic stability of the superconducting state in the grain and Josephson coupling between the grains. The inter-grain superconductivity is known to exhibit any magnetic anomaly in the third harmonic susceptibility [2,5]. We are trying to detect the quite small third harmonic response, whose magnitude is two or three orders lower than the first harmonic one, in extremely strict experimental environment using DAC.

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