

# Critical temperature oscillation in the thermal cycle below 16 K in $\text{Y}_{0.83}\text{Ca}_{0.17}\text{Ba}_2\text{Cu}_3\text{O}_6$

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## Abstract

In an  $\text{Y}_{0.83}\text{Ca}_{0.17}\text{Ba}_2\text{Cu}_3\text{O}_6$  polycrystalline sample, under thermal cycling below 16 K, an unusual relaxation effect of the superconducting resistive transition has been observed after the pressure is changed at  $RT$ . The normal-state resistivity does not change. Since there is no mobile oxygen in the present sample, the effect of oxygen rearrangement can be ignored. The observed results can be understood by the charge redistribution within the  $\text{CuO}_2$  planes.

*Key words:* granular superconductivity; pressure effect; Josephson coupling; charge redistribution

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## 1. Introduction

For some high- $T_c$  cuprates, such as  $\text{YBa}_2\text{Cu}_3\text{O}_{6+y}$  (Y123) with  $\text{CuO}_y$  chain and  $\text{Tl}_2\text{Ba}_2\text{CuO}_{6+\delta}$  (Tl2201) with  $\text{Tl}_2\text{O}_{2+\delta}$  layer, pressure can induce oxygen rearrangement (OR) within these charge reservoir layers, which significantly enhances the pressure effect on the critical temperature  $T_c$  and Hall coefficient  $R_H$  [1,2]. Since the oxygen configuration is frozen at the low temperature ( $LT$ ), typically below  $\sim 100$  K, the pressure effect is substantially reduced by changing pressure at  $LT$ . This procedure has led to the observation of relaxation effect that the pressure effect on  $T_c$  or  $R_H$  due to pressure change at  $LT$  eventually evolves into that at  $RT$  due to thermally activated OR [1,3].  $\text{Y}_{1-x}\text{Ca}_x\text{Ba}_2\text{Cu}_3\text{O}_6$  is a system without  $\text{CuO}_y$  chain and therefore free from the OR effects under pressure [2]. Ca doping destroys the antiferromagnetic phase and incipient superconductivity signaled by a resistive kink at  $T_{c1}$  is observed at  $x=0.17\text{--}0.18$  and a zero-resistivity state at  $\sim 0.20$  [4]. This is consistent with microscopic electronic phase separation into hole rich region and hole poor one observed by the muon spin relaxation experiment [5]. In the "local" supercon-

ductor  $\text{Y}_{0.83}\text{Ca}_{0.17}\text{Ba}_2\text{Cu}_3\text{O}_6$ , we have observed an unexpected pressure-induced two-step transition at  $T_{c2}$  and  $T_{c3}$  [6]. Further, a relaxation effect at  $RT$  has been observed for the pressure range between 0.1 and 0.3 GPa. The two-step transition exhibits strong current effect [6]. Therefore, the superconductivity at  $T_{c1}$  is attributed to the superconducting islands, and those at  $T_{c2}$  and  $T_{c3}$  are attributed to the Josephson coupling between the islands. Since no pressure effect on  $R_H$  is observed [2], both results could come from the pressure-induced charge redistribution within the  $\text{CuO}_2$  planes. In this paper, we have studied the  $LT$  thermal cycling effect on  $\rho(T)$  or  $T_c$  of a sample quenched to below 85 K right after releasing the pressure from 0.35 GPa to 0.15 GPa at  $RT$ .

## 2. Experimental

$\text{Y}_{0.83}\text{Ca}_{0.17}\text{Ba}_2\text{Cu}_3\text{O}_6$  was prepared by a solid state reaction [2]. Pressure was changed at  $RT$  by a piston cylinder method, and determined by the Pb manometer. The pressure medium was a mixture of flourinert (FC77:FC70=1:1). The resistivity  $\rho$  was measured by ac four-probe method. Temperature  $T$  was determined

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by the k-type thermocouple or Ge temperature sensor.

### 3. Results and discussion

After the pressure was released from 0.35 GPa to 0.15 GPa at  $RT$ , the sample was quenched below 85 K. In such low-pressure range, the solidification effect of pressure medium can be ruled out. The  $\rho(T)$  measurement was carried out under the thermal cycling between 1 K and 85 K to exclude the influence of the unwanted *OR*. The Pb manometer always indicated 0.15 GPa. When the sample was finally heated up to  $RT$ , the  $\rho(T)$  (open circles) was identical to that in the first heating process (closed circles), except for the behavior below  $T_{c2}$  as shown in the inset of Fig. 1. Accordingly, the  $\rho(T > T_{c2})$  is found not to be influenced by the *LT* thermal cycle. A significant change in  $\rho(T < T_{c2})$  was induced by the thermal cycling below 16 K. The typical  $\rho(T)$  is shown in Fig. 1. Below  $T_{c2}$ , the  $\rho(T)$  decreases in two steps with reducing  $T$  (curve1). Subsequent thermal cycling brings a significant change in  $\rho(T)$  around the tail part of the transition at  $T_{c2}$ . As the *LT* thermal cycling proceeds, the transition at  $T_{c2}$  has a shorter tailing part and becomes sharper. The  $\rho(T)$  below  $\sim 3$  K eventually increases with decreasing  $T$  and the transition at  $T_{c3}$  disappears. The insulating like  $\rho(T)$  below  $\sim 3$  K appears instead of the transition at  $T_{c3}$ . As the transition at  $T_{c2}$  is enhanced, that at  $T_{c3}$  is suppressed. This suggests a charge redistribution among the superconducting regions responsible for the transition at  $T_{c2}$  and that at  $T_{c3}$ .

We summarize the evolution of the oscillation of  $\rho(T < T_{c2})$  due to *LT* thermal cycling as a function of time in Fig. 2. The time zero is defined as the time right after the release of pressure at  $RT$  and each pair (closed and open circles) of data points represents one thermal cycling between 1.3 and 65-85 K until 285 hr and between 1.3 and 16 K over 285 hr. The  $\rho_{7K}$  and  $\rho_{2K}$ , resistivity at 7 K and 2 K respectively, for the curve 1-4 in Fig. 1 are in the time domain of 320 - 416

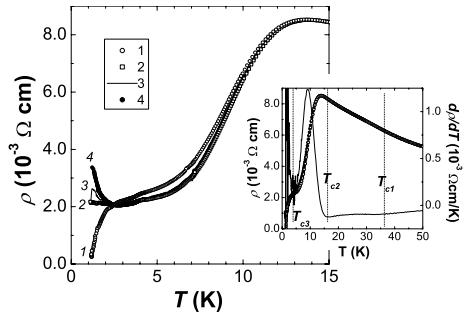


Fig. 1.  $\rho(T)$  under the thermal cycling below 16 K. The inset shows the  $\rho(T)$  until 70 K. The excitation current was 30  $\mu$ A.

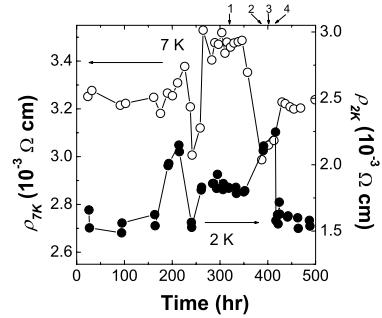


Fig. 2.  $\rho_{7K}$  (open cycles) and  $\rho_{2K}$  (closed cycles) as a function of the time. The number corresponds to curves shown in Fig. 1.

hr. As the *LT* thermal cycling proceeded,  $\rho_{7K}$  and  $\rho_{2K}$  were found to oscillate. Until  $\sim 260$  hr, both changed with the same trend. But, over  $\sim 260$  hr, the trend in  $\rho_{7K}$  was opposite to that in  $\rho_{2K}$ . The result over  $\sim 260$  hr suggested a charge redistribution through quantum tunneling between two electronic states. Beyond  $\sim 260$  hr, the electronic states of the sample were tuned by the *LT* thermal cycling.

The oscillation in  $\rho(T < T_{c2})$  is observed in the cooling process after the change in pressure at  $RT$ , in strong contrast to the usual relaxation effect observed in Y123 or Tl2201 [1]. This suggests no possibility of the *OR* and this oscillation is purely electronic in nature. Since the charge distribution varies with time in spite of a frozen local structure, the superconductivity may be responsible to the variation of the electronic states. Indeed, this seems to be consistent with our observation of an intrinsic electronic phase separation of doped holes in the  $\text{CuO}_2$  planes into distinct superconducting phases [7].

In summary, we have observed the unexpected oscillation of  $\rho(T)$  below  $T_{c2}$ . This is characteristic different from the normal relaxation effect on  $T_c$  through the *OR*. The present result can be explained by the charge redistribution within the  $\text{CuO}_2$  planes

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