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# Field–Temperature Phase Diagram of Intergrain Ordering in Superconducting Ceramic YBCO

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# Field–Temperature Phase Diagram of Intergrain Ordering in Superconducting Ceramic YBCO

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**Abstract**. The ceramic $YBa_2Cu_4O_8$ superconductor composed of submicron grains is considered a random Josephson-coupled network containing the so-called $\pi$ junctions and shows successive phase transitions. With decreasing temperature, first the intragrain superconductive transition occurs inside each grain at $T_{c1}$ and then the chiral-glass transition occurs among the grains at $T_{c2}$ (< $T_{c1}$). The third transition at $T_{c3}$ (< $T_{c2}$) is the intergrain superconducting transition. We measured the nonlinear susceptibility and resistivity of the ceramic $YBa_2Cu_4O_8$ superconductor to determine the field dependences of the transition temperatures $T_{c2}$ and $T_{c3}$. The phase diagram of the intergrain ordering is discussed in light of the result predicted by Kawamura.

## 1. Introduction

Ceramic cuprate superconductors composed of submicron grains are considered as random Josephson-coupled networks of 0 and $\pi$ junctions. The circulation of a local loop-supercurrent and an orbital moment are generated spontaneously when there is an odd number of $\pi$ junctions in a closed loop ($\pi$ loop). The frustration effect due to the random distribution of $\pi$ loops should lead to the chiral-glass state predicted by Kawamura and Li.[1] Kawamura investigated the ordering of ceramic cuprate superconductors, which is well described by the isotropic 3D *XY* spin-glass model.[2-3] Namely, the spin in a magnet corresponds to the phase of the superconducting order parameter at each grain, while the chirality corresponds to the circulation of superconducting current loops flowing in a Josephson-coupled network. At the chiral-glass transition, the chirality is frozen, i.e., the circulation of superconducting current loops is frozen, which can be observed experimentally as a negative divergence of the nonlinear susceptibility. At the spin-glass transition, the phase of the superconducting order parameter is frozen and the system becomes a true superconductor. This can be observed experimentally as the onset of zero resistivity. Kawamura predicted that successive chiral- and spin-glass transitions occur at $T = T_{CG}$ and $T = T_{SG}$ with $T_{CG} > T_{SG}$ in an isotropic system. [2] It is interesting to check experimentally whether the spin-chirality decoupling phenomenon is realized in the ordering of ceramic cuprate superconductors. Through experimental studies for ceramic $YBa_2Cu_4O_8$, the critical temperature

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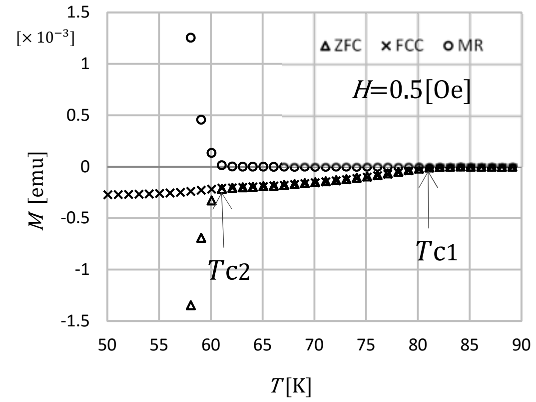


**Figure 1.** Temperature dependence of the zero-field magnetization (ZFC), field-cooled magnetization (FCC), and thermoremanent magnetization (MR) at $H$ = 0.5 Oe

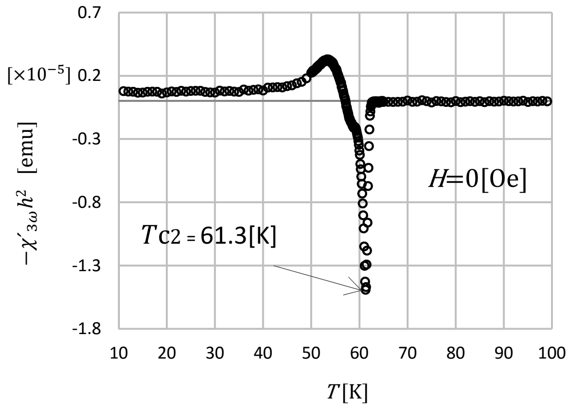


**Figure 2.** Temperature dependence of the nonlinear susceptibility at 1.0 Hz with the ac field amplitude of 0.1 Oe at $H$ = 0 Oe

$T_{c2}$ of the chiral-glass transition was detected using the nonlinear susceptibility [4-5] and the critical temperature $T_{c3}$ of the intergrain superconductive transition was estimated using the onset of the zero linear resistivity at zero field.[6-7] In the chiral-glass phase below $T_{c2}$, the magnetic glass behavior and memory effects, similar to those of spin-glasses, were observed in ceramic $YBa_2Cu_4O_8$.[8-9]

In this work, we investigated the field dependence of $T_{c2}$ and $T_{c3}$ of ceramic $YBa_2Cu_4O_8$ and discussed the field–temperature phase diagram of the intergrain ordering.

## 2. Sample and experiments

The sample was prepared using the citrate pyrolysis method.[10] The precursor was calcined for 100 h at 777 °C to yield a pure $YBa_2Cu_4O_8$ phase, which was sieved and pressed, and then sintered for 85 h at 780 °C, to became a pure $YBa_2Cu_4O_8$ ceramic with submicron grains. The dc magnetization and the ac susceptibility were measured with a SQUID magnetometer (Quantum Design MPMS-5). The linear resistivity was measured by low-level pulsed electrical characterization with a current source (KEITHLEY 6221) and a nanovoltmeter (KEITHLEY 2182A) combination. The delta method for low voltage measurements was used to eliminate the constant thermoelectric voltage and to minimize the amount of power dissipated in the sample. An external field was applied perpendicular to the dc current.

## 3. Results

We measured the temperature dependences of the dc magnetization and ac susceptibility to determine the transition temperatures $T_{c1}$ and $T_{c2}$. Figure 1 shows the temperature dependences of the zero-field-cooled, field-cooled, and thermoremanent magnetizations at $H$ = 0.5 Oe. The transition at $T_{c1}$ = 81 K was identified as the intragrain superconducting ordering, in which small diamagnetism due to the Meissner effect appears in zero-field-cooled and field-cooled magnetizations. The zero-field-cooled magnetization steeply branches away from the field-cooled one at the glass transition temperature $T_{c2}$ = 61 K. This branching temperature coincides with thermoremanent magnetization vanishing point, which must be caused by the release of magnetic flux trapped in the ordered network.

The nonlinear susceptibility estimated from the first term of the series of in-phase odd-harmonic responses at 1.0 Hz with an ac field amplitude of 0.1 Oe is shown in Figure 2. A negative peak of nonlinear susceptibility was observed at $T_{c2}$ = 61.3 K; at this temperature, the ceramic $YBa_2Cu_4O_8$ underwent a chiral-glass transition at zero applied field. We measured the temperature dependence of nonlinear susceptibility at some applied fields of magnitude up to 300 Oe and estimated $T_{c2}$ from the peak temperature of nonlinear susceptibility in applied fields. The negative peak of nonlinear

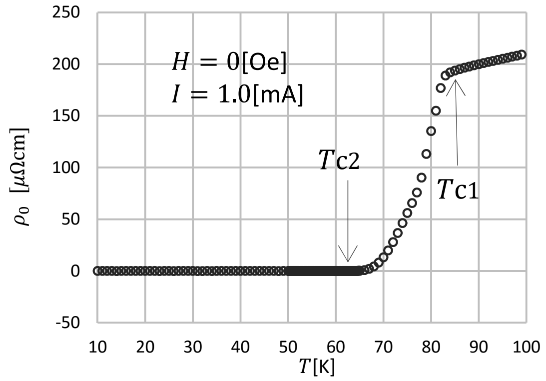


**Figure 3.** Temperature dependence of the linear resistivity for the applied dc current of 1.0 mA at $H = 0$ Oe

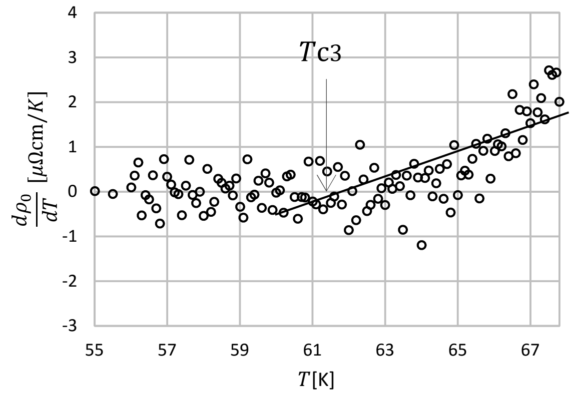


**Figure 4.** Temperature dependence of $d\rho_0/dT$ for the applied dc current of 1.0 mA at $H = 0$ Oe

susceptibility shifts slightly toward higher temperatures with increasing the field magnitude up to 30 Oe, and the magnitude of the peak gradually increases. However, the peak on the high-field side shifts remarkably toward lower temperatures with increasing the field magnitude up to 300 Oe and the magnitude of the peak remains almost constant. The field dependence of $T_{c2}$ at low fields is not normal in chiral-glass ordering. In the fully isotropic Heisenberg case, the chiral-glass transition temperature should be independent of the field magnitude and should be constant at low fields. [2]

Next, we measured the temperature dependence of the linear resistivity $\rho_0$ for the applied dc current of 1.0 mA at some applied fields up to 100 Oe. Figure 3 shows the temperature dependence of $\rho_0$ at zero field. As the temperature was reduced, $\rho_0$ decreased rapidly at $T_{c1}$, and then decreased monotonously and almost disappeared around $T_{c2}$. It is difficult to estimate the transition temperature $T_{c3}$ for $\rho_0 = 0$ because $\rho_0$ diminishes continuously with decreasing temperature. Therefore, $T_{c3}$ was estimated as the temperature at which $d\rho_0/dT = 0$, as shown in Figure 4. At zero field, the intergrain superconducting transition temperature was identified as $T_{c3} = 61.8$ K; this temperature is close to the chiral-glass temperature $T_{c2} = 61.3$ K at zero field. The critical temperature $T_{c3}$ shifts remarkably to the lower temperature side with increasing field magnitude and decreases 59.8 K at $H = 100$ Oe. The change and field dependence of $T_{c3}$ in applied fields are very different from those of $T_{c2}$.

## 4. Discussion and conclusions

The transition temperatures $T_{c2}$ and $T_{c3}$ were obtained at zero field and some applied fields. Figure 5 shows the field–temperature phase diagram for the ceramic $YBa_2Cu_4O_8$. The diagram is divided into three phases, namely, the intragrain superconducting phase, the chiral-glass phase, and the intergrain superconducting phase, by $T_{c2}$ and $T_{c3}$.

At zero field, chiral-glass ordering takes place at the same temperature as intergrain superconducting ordering ($T_{c2} = T_{c3}$). This means that the chirality is not decoupled from the phase of the superconducting order parameter (corresponding to the spin in a spin-glass system) in the ceramic $YBa_2Cu_4O_8$ at zero field. However, when a field is applied, both the temperature value and the field dependence of $T_{c2}$ are different from those of $T_{c3}$. Thus, "spin-chirality decoupling" occurs in an applied field in the ceramic $YBa_2Cu_4O_8$.

Kawamura predicted that the "spin-chirality decoupling" phenomenon is realized in the ordering of granular cuprate superconductors in zero and applied fields.[2] The effects of magnetic fields on the spin-glass ordering at $T_{SG}$ (corresponding to $T_{c3}$ in the ceramic $YBa_2Cu_4O_8$) and on the chiral-glass ordering at $T_{CG}$ (corresponding to $T_{c2}$ in the ceramic $YBa_2Cu_4O_8$) were studied by Kawamura.[2] According to the Kawamura, the spin-glass transition line should exhibit a singular form, $|T_{SG}(0) - T_{SG}(H)|^2 \propto H$ . We estimated the exponent from the field dependence of $|T_{c3}(0) - T_{c3}(H)|$ at high fields.

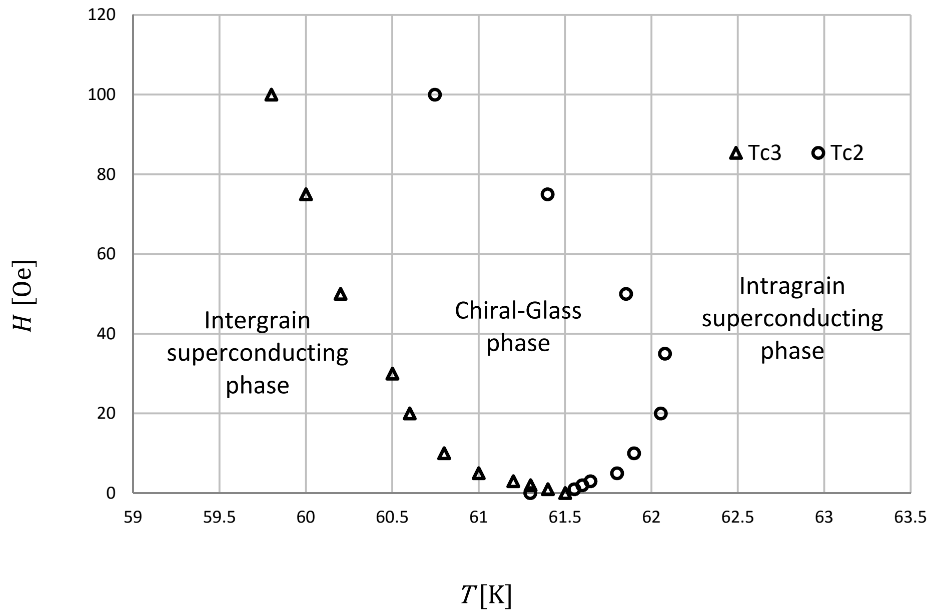


**Figure 5.** Phase diagram showing intergrain superconducting, chiral-glass, and intragrain superconducting phases as a function of temperature and field in the ceramic $YBa_2Cu_4O_8$. Triangle and circles correspond to $T_{c3}$ and $T_{c2}$, respectively.

The estimated value of 2.3 is within the range expected from the theoretical value of 2. The chiral-glass transition temperature $T_{CG}(H)$ under applied fields should behave as the so-called the Gabay-Toulouse (GT) line of the mean-field model, $|T_{CG}(0) - T_{CG}(H)| \propto H^{1/2}$.[11] At a field magnitude lower than $H$= 30 Oe, $T_{c2}$ increases gradually with increasing field magnitude, which is different from the field dependence of the GT line. However, the obtained exponent from the field dependence of $|T_{c2}(0) - T_{c2}(H)|$ at high fields is 0.53, which is consistent with the theoretical value of 1/2. Therefore, the transition lines of $T_{c2}(H)$ and $T_{c3}(H)$ at high fields seem to correspond with the chiral-glass and the intergrain superconducting transition lines, respectively. In an applied field, the chirality is decoupled from the phase of superconducting order and chiral-glass ordering takes place at a temperature higher than intergrain superconducting ordering, $T_{c2}(H) > T_{c3}(H)$.

In conclusion, we investigated the temperature dependences of nonlinear susceptibility and linear resistivity in zero and applied fields, and estimated the transition temperatures $T_{c2}$ and $T_{c3}$ at zero and some applied fields in the ceramic $YBa_2Cu_4O_8$. The field–temperature phase diagram of the intergrain ordering was obtained and compared with the expected phase diagram from the chirality scenario described by Kawamura. The results suggest that spin-chirality decoupling takes place at finite fields in granular cuprate superconductors.

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