Magnetic Memory Effects in a Chiral-Glass Phase of a Superconductive Ceramic YBa$_2$Cu$_4$O$_8$

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Abstract. The ceramic YBa$_2$Cu$_4$O$_8$ composed of sub-micron size grains is considered as random Josephson-coupled networks of 0 and $\pi$ junctions and shows successive phase transitions. The upper transition occurs inside each grain at $T_{c1}$ and the lower transition occurs among the grains at $T_{c2} (< T_{c1})$, where a negative divergence of nonlinear susceptibility is found. This critical phenomenon at $T_{c2}$ suggests the onset of the chiral-glass phase predicted by Kawamura and Li. We investigated the magnetic memory phenomena in the temperature dependence of thermoremanent magnetization (TRM). The memory effects are observed in TRM measured on heating after the cooling process with a halt at the temperature $T_s$ below $T_{c2}$. The results indicate the existence of a glassy phase such as the chiral-glass one.

1. Introduction

Ceramic high-$T_c$ superconductors, which are composed of homogeneous sub-micron-size grains, may be regarded as random Josephson-coupled networks that contain so-called $\pi$ junctions. The circulation of a local loop-supercurrent is generated spontaneously in a zero field when there is an odd number of $\pi$ junctions in a closed loop that consists of some Josephson junctions. Theoretically it is suggested that these spontaneous current loops are affected by an external magnetic field and a net positive magnetization is produced.[1] Furthermore, the frustration effect due to the random distribution of $\pi$ junctions should lead to the chiral-glass state predicted by Kawamura and Li.[2] They investigated the chiral-glass phase of ceramic high-$T_c$ superconductors using Monte Carlo simulations. In experimental studies, ceramic high-$T_c$ superconductors such as YBa$_2$Cu$_3$O$_{6.9}$,[3,4], YBa$_2$Cu$_3$O$_{7.6}$[5] and Bi$_2$Sr$_2$CaCu$_2$O$_{8}$[6] samples has been investigated by several magnetic and transport measurements. The critical phenomenon at the transition temperature and the collective behavior in the glass state of these real samples suggest the existence of the chiral-glass phase.

In the ceramic samples, the magnetic memory behavior was investigated only in Bi$_2$Sr$_2$CaCu$_2$O$_8$ one.[7,8] The memory effects in Bi$_2$Sr$_2$CaCu$_2$O$_8$ samples were observed in the zero-field-cooled (ZFC) and TRM magnetization vs. temperature curves measured on heating after certain cooling.
protocols. However, the sample is a strong Josephson-coupled system, and the transition temperature of inter-grain superconductivity is close to that of intra-grain glass phase. Therefore, it is difficult to distinguish between the intra-grain and inter-grain ordering properties. The ceramic YBa$_2$Cu$_4$O$_8$ and YBa$_2$Cu$_3$O$_7$-$\delta$ have shown successive superconducting transitions under zero field. With decreasing temperature, a transition occurs at first inside each grain at $T_{c1}$ and the second transition occurs among the grains at $T_{c2}$. The discrepancy between the field–cooled (FC) and ZFC magnetization levels appears at below $T_{c2}$, and negative divergence of nonlinear susceptibility is observed at $T_{c2}$, which may reflect a chiral-glass transition.\cite{9} In such extremely weak-Josephson-coupled system, the transition in the intra-grain and inter-grain regions are well separated in temperature. In the present work, we investigate the magnetic memory effects for the typical weak-link system of the ceramic YBa$_2$Cu$_4$O$_8$ in order to clarify the properties of the glass phase.

2. Sample and experiments
The sample of ceramic YBa$_2$Cu$_4$O$_8$ is synthesized by the procedure reported in Ref.\cite{10}. The sub-micron powder was prepared using the citrate pyrolysis method. The precursor was calcined for $120 \, \text{h}$ at $778 \, ^\circ\text{C}$ to yield the pure YBa$_2$Cu$_4$O$_8$ phase, which was sieved and pressed, and then sintered for $50 \, \text{h}$ at $780 \, ^\circ\text{C}$. The dc magnetization and the ac susceptibility were measured with a SQUID magnetometer (Quantum Design MPMS-5) using the ultra-low-field option and the temperature sweep operation. The sample space in the magnetometer was shielded with $\mu$-metal. As a result, the residual field was reduced to less than $10 \, \text{mG}$. The nonlinear susceptibility was derived from the harmonics in-phase Fourier component for the ac field response.

3. Results
We measured the temperature dependence of the dc magnetization and ac susceptibility, to determine the transition temperatures $T_{c1}$ and $T_{c2}$. Figures 1 and 2 show the temperature dependence of the ZFC, FC and TR magnetizations at $H = 0.1 \, \text{G}$. The upper transition at $T_{c1}=81 \, ^\circ\text{K}$ was identified as the inter-grain superconducting ordering, in which the small diamagnetism due to the Meissner effect appears in ZFC and FC magnetizations. The TRM and the discrepancy between the FC and ZFC magnetizations appeared at around $54 \, \text{K}$. Below $54 \, \text{K}$, the equality $M_{\text{ZFC}} - M_{\text{FC}} = M_{\text{TRM}}$ is closely satisfied for all temperatures.

The linear susceptibilities, in-phase susceptibility $\chi'$ and out-of-phase one $\chi''$, are measured in the frequency of $1.0 \, \text{Hz}$ with an ac-field amplitude of $0.1 \, \text{G}$ under the zero external field. As shown in Figure 3, $\chi'$ changes abruptly at around $54 \, ^\circ\text{K}$ and the behavior is similar to that of $M_{\text{ZFC}}$. $\chi''$ has a maximum at around $53 \, ^\circ\text{K}$ and exhibits strong dissipation even in low-frequency region. The
nonlinear susceptibility $\chi_2$ estimated from the first three terms of the series of in-phase odd-harmonic responses is shown in Figure 4. Negative divergence of $\chi_2$ was observed at $T_{c2}=54.5$ K, at which temperature the ceramic YBa$_2$Cu$_4$O$_8$ underwent a chiral-glass transition.

The magnetic memory experiments are performed using the temperature dependence of TRM. The sample was rapidly cooled (cooling rate of 3.5 K/min) in $H = 0.1$ G from 90 K above $T_{c1}$ down to a temperature $T_s$ below $T_{c2}$, at which a halt was made for $t_w=10000$ s. The cooling was then resumed down to 30 K with cooling rate of 3.5 K/min, and the magnetic field is turned off. The TRM was then recorded on slow heating process (heating rate of 0.14 K/min). We have measured TRM experiments with the fixed wait time $t_w=10000$ s for three stop temperatures $T_s=52$, 47, and 42 K. Figure 5 shows the influence of a temperature stop of $t_w=10000$ s at $T_s = 52$ K close to $T_{c2}$ on the TRM. The TRM plots corresponding to the stop experiment around $T_s$ lies slightly above the reference plots measured with the same cooling and heating rates. Figure 6 shows the difference $\Delta M_{TRM}=M_{TRM}(T_{s}=52)$ K in Figure 5. The influence of the halt is confined to a narrow temperature region near the halt temperature $T_s$. The state becomes frozen in for further cooling down to the lowest temperature, and is retrieved on reheating. Similar memory effects were observed in the ceramic Bi$_2$Sr$_2$CaCu$_2$O$_8$ samples [7,8] and also in disordered magnetic systems such as spin glasses.[11,12] The Bi$_2$Sr$_2$CaCu$_2$O$_8$ sample is a strong Josephson-coupled system, so the transition temperature of inter-grain superconductivity is close to that of intra-grain one. Therefore, it is difficult to distinguish the memory effects due to the intra-grain or those due to inter-grain superconducting transition. Also, the nonlinear susceptibility does not diverge at the transition temperature.

The ceramic YBa$_2$Cu$_4$O$_8$ is considered as a typical weak-link system. The inter-grain transition at $T_{c2}=54.5$ K separate well from the intra-grain transition at $T_{c1} = 81$ K in temperatures. Also the nonlinear susceptibility shows the negative divergence at $T_{c2}$. The memory phenomena of TRM are observed below $T_{c2}$. The peak of $\Delta M_{TRM}$ is large in the case for $T_s = 52$ K near the temperature at the
maximum of $\chi''$ and becomes small in the case for $T_s = 42$ K where $\chi''$ is close to zero. The observation of $\chi''$ at low frequency shows the existence of a slow dynamics near the glass-transition. The origin of the memory effects may relate to the slow dynamics in chiral-glass phase. The results suggest that chiral-glass ordering occurs at $T_{c2}$ in the ceramic YBa$_2$Cu$_4$O$_8$.

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