Memory effects in $YBa_2Cu_3O_{7-\delta}$ single crystal

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Measurements of the time dependence of zero-field-cooled (ZFC) and field-cooled (FC) magnetization M in $YBa_2Cu_3O_{7-\delta}$ single crystal have been performed as a function of temperature and magnetic field. The appearance of an echo-like feature in the decay rate $S = dM/d \ln t$ at an observing time t equal to the waiting time t_w during which the specimen was prepared at a given field H_0 and temperature T reveals aging effects in the superconducting state. Similar phenomena reported in spin glasses seem to validate the picture of a superconducting glass state in $YBa_2Cu_3O_{7-\delta}$.

1. Introduction

In the early stages of discovery of the high-T_c superconducting oxides, Müller et al. [1] pointed out the irreversible nature of the magnetization, and in particular the difference between the magnetic responses of field-cooled (FC) and zero-field-cooled (ZFC) samples. The existence of a superconducting glass state was proposed; its origin was suggested to arise from the extremely short coherence length ξ (3–30 Å) found in these materials [2]. The superconducting order parameter at the superconducting region boundaries is depressed, and proximity-effect junctions are created at twin boundaries in $YBa_2Cu_3O_{7-\delta}$ or at other planar defects. Such a network of weak links can produce the observed glassy properties proposed by theory [3] and numerical simulations [4]. Many experiments have been performed to check these glass-like properties. Logarithmic time decay of the magnetization M was reported by several groups and was measured as a function of temperature and magnetic field in ceramics [5, 6], single crystals [7–9], and thin films [10]. Although suggestive of a glass state, the time dependence of M was also proposed to originate from giant flux creep [9] specific to the oxides, because of the much lower activation energy needed to depin the flux lines as compared to conventional superconductors.

We have measured the time dependence of M as a function of T and H in a single crystal of $YBa_2Cu_3O_{7-\delta}$ either FC or ZFC below T_c. A surprising observation is that the decay rate $S = dM/d \ln t$ is not uniquely determined, but depends on the waiting time t_w during which the specimen has been prepared in a certain (T, H_0) state before applying a field-change ΔH and starting the relaxation measurement. The echo-like or memory feature appearing as an inflection point in the decay of M vs. $\ln t$ shows that the system ages through multiple relaxation processes over a hierarchy of free-energy barriers. Similar effects have been discovered in spin glasses and extensively studied both experimentally [11] and theoretically [12]. We propose that our data in $YBa_2Cu_3O_{7-\delta}$ support the picture of a glass state owing to the presence of frustrated superconducting domains, in particular close to T_c.

2. Experiment

A thin single-crystal platelet with the c-axis perpendicular to its plane was mounted in a SHE SQUID susceptometer with the magnetic field applied parallel to c. Great care was taken to check and ensure the field and temperature stability over long-term runs in order to eliminate any spurious effects. The contribution of the sample holder was negligible compared to the diamagnetic signal of the specimen in the temperature range used.

The magnetization was measured vs. T in fields up to 200 Oe and showed well-defined FC and ZFC branches with the onset of superconduction at $T_c = 92$ K. The good superconducting properties of the sample were further verified by measuring at 5 K the hysteresis curve of M vs. H between 0 and 4 tesla. The experimental procedure is summarized in **Figure 1**. As the specimen reaches the temperature $T_m < T_c$ after the cooling process in zero field (1) or in a field H_0 (2), the clock measuring its aging time t_a is started. The system is left in this quasi-equilibrium state for a certain waiting time t_w. Its magnetization is then modified by applying a field perturbation ΔH and its decay measured, starting at time $t = 0$ (or $t_a = t_w$). The observation time t is related to the aging time by $t = t_a - t_w$.

3. Results and discussion

Initially the relaxation measurements were done with almost no waiting time ($t_w \approx 0$). The temperature dependence of the logarithmic decay rate $S = dM/d \ln t$ found in the ZFC case ($\Delta H = 1$ kOe) shows a broad maximum around 30 K (**Figure 2**). The initial increase of $S(T)$ at low T and its asymptotic decrease close to T_c have been reported by several groups [6, 8–10]. In the FC case, the subsequent application of ΔH leads to almost the same amplitude and T-dependence of S; however, the rate S measured under the same FC conditions but without applying ΔH is about 1000 times smaller and of opposite sign. It is almost temperature-independent below 50 K, then rises to peak around 60–70 K before vanishing at T_c, in accordance with [9].

In **Figure 3** the decay of the ZFC magnetization M measured at 75 K is displayed on a logarithmic time scale for three different waiting times $t_w = 1$ h, 6 h, and 24 h. The value of ΔH is 1 kOe. H_0 is not exactly zero, but has the value of the remanent field of the superconducting coil (about 20 Oe, as checked later).

Aside from the initial transient response at very short times, possibly due to fast reversible relaxation processes [13], M decays as $\ln t$. However, at times equivalent to the waiting time t_w (vertical arrows), the decay rate increases suddenly, as shown by the systematic deviation from the $\ln t$ behavior. This inflection point, partly hidden for longer waiting times t_w (e.g., 24 h) because of the saturation of M toward equilibrium, is therefore the echo-like response to the field step applied at $t = 0$. This effect is better seen when the sample is really cooled in a larger field H_0. The frustration between the superconducting domains may be better established, yielding a larger hierarchy of relaxation processes. The inflection point around t_w in the decay of M vs. $\ln t$ is indeed sharper for $H_0 = 0.5$ kOe, as demonstrated

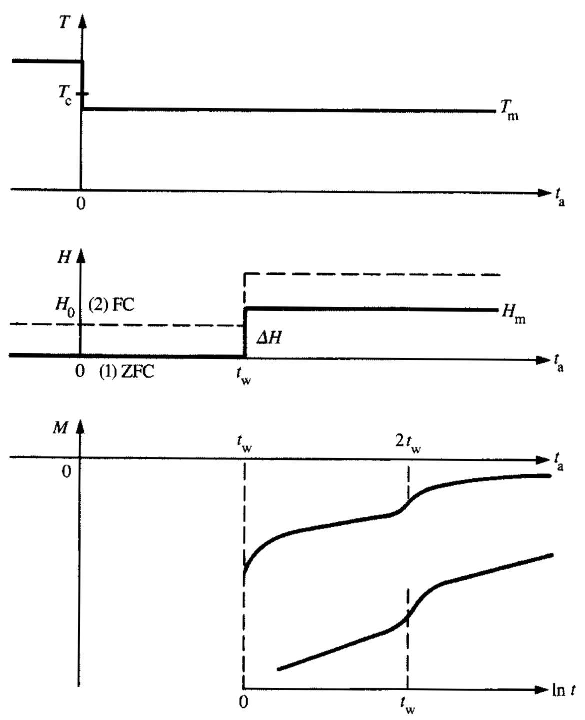

Figure 1

Measurement procedure of M vs. t. The sample is either zero-field-cooled (1) or field-cooled (2) to $T_m < T_c$ and left in that state for a waiting time t_w before applying a field step ΔH at time $t = 0$ and starting the relaxation measurement of the magnetization M. The aging time of the system is $t_a = t_w + t$.

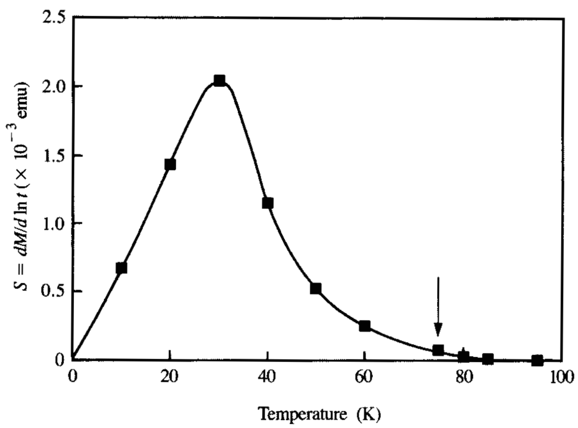

Figure 2

Magnetization decay rate S vs. temperature T of Y–Ba–Cu–O single crystal cooled in zero field. The solid line is a guide to the eye. The arrow indicates $T = 75$ K, where memory effects were measured.

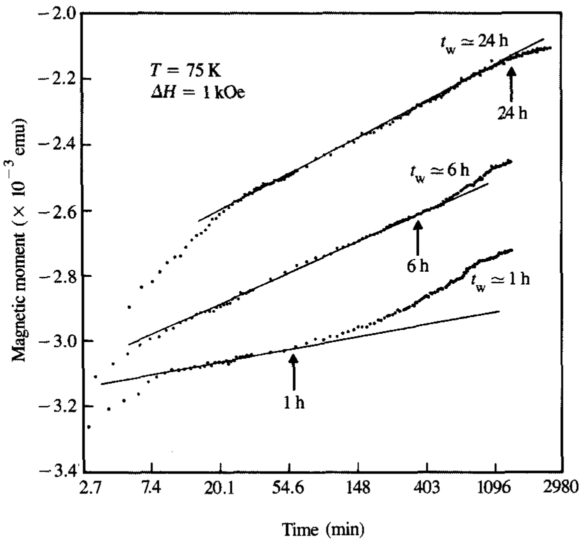

Figure 3

Magnetic moment M vs. ln t of the ZFC Y–Ba–Cu–O single crystal measured at 75 K after waiting either t_w = 1 h, 6 h, or 24 h. ΔH is 1 kOe. For clarity, the curves with t_w = 1 h and 24 h have been shifted down by 6×10^{-4} emu and up by 4×10^{-4} emu, respectively.

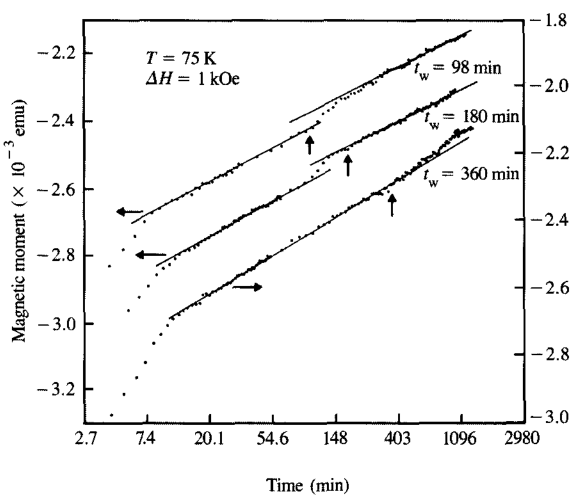

Figure 4

Magnetic moment M vs. ln t of the FC Y–Ba–Cu–O single crystal measured at 75 K after waiting t_w = 98 min, 180 min, and 360 min in H_0 = 0.5 kOe. ΔH is 1 kOe.

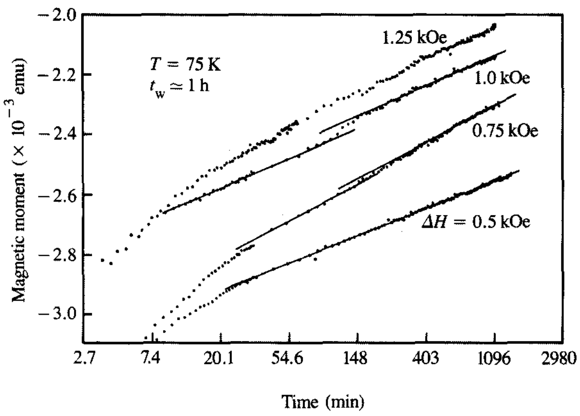

Figure 5

Effect of the field jump amplitude ΔH = 0.5, 0.75, 1.0, 1.25 kOe on the occurrence of the inflection point in M vs. ln t for the FC Y–Ba–Cu–O single crystal at 75 K. H_0 = 0.5 kOe and $t_w \approx$ 1 h.

in **Figure 4**. In that case t_w = 98 min, 180 min, and 360 min were used. The fact that the echo-like jump occurs sometimes at a time slightly different from t_w is possibly due to some uncontrollable fluctuations in reaching the initial state (T_m, $H_m = H_0 + \Delta H$) at $t = 0$. Not only the H_0 amplitude, but also the value of ΔH seems critical. It is seen in **Figure 5** that for the sample prepared under the same conditions (T = 75 K, H_0 = 0.5 kOe, and $t_w \approx$ 1 h), the characteristic feature in M vs. ln t is only visible for ΔH between ~0.75 and 1 kOe. This result qualitatively agrees with the H–T phase diagram of the superconducting glass state proposed by the model of Morgenstern [4]. Furthermore, memory effects seem to be more readily observable in the temperature range $0.7 \leq T/T_c \leq 0.9$, where the decay rate of the FC magnetization ($\Delta H = 0$) is observed to be larger.

In order to explain our data, we suggest that the magnetic field which penetrates the superconductor induces relaxation processes through the movement of vortices between and within weakly coupled superconducting domains. These domains of different sizes may grow with time as the system tends to reach equilibrium. The origin of the relaxation processes is not quite clear in complicated systems like the oxides. It is nevertheless expected that the large movement of vortices is favored by anomalously weak pinning strengths in these materials. A simple picture for describing the occurrence of the echo or memory effect observed here may be given by following the time evolution of the density of relaxation processes $D(\tau, t_a, H)$ which remain active during the aging of the system (**Figure 6**). At time $t_a = 0$, the initial distribution has a given shape defined by $g_H(\tau)$ which goes to zero for $\tau > \tau_{max}$. The time dependence of D is expressed by $D(\tau, t_a, H) = g_H(\tau) \exp(-t_a/\tau)$. Such an "off-equilibrium" distribution, normalized to a horizontal line in Figure 6, is similar to a distribution of barrier heights U which characterizes the free-energy landscape in which the system is constrained. As the FC superconductor is left in its (T_m, H_0) state for a time t_w, the short-time processes relax so that $D_1(\tau, 0, H_0)$ evolves into $D_1(\tau, t_w, H_0)$. The application of a field step ΔH at $t_a = t_w$ adds a second distribution $D_2(\tau, t_w, H_m)$ whose short-time components ($\tau \le t_w$) vanish first. ΔH should not be too large, to avoid washing out the initial distribution D_1, which continues to evolve but now at a much smaller rate than D_2 (on a logarithmic time scale). As the aging time t_a approaches $2t_w$ (or $t \approx t_w$), the processes of D_1 with $t > t_w$ contribute progressively more to the decay of the magnetization. Therefore, the measured dependence of M vs. ln t with its inflection point at $t \approx t_w$ is expected to follow the integrated shape of the distribution $D(\tau, 2t_w, H_m) = D_1 + D_2$ shown as curve (5) in Figure 6.

In conclusion, we argue that the aging effects in $YBa_2Cu_3O_{7-\delta}$ with echo-like features in M vs. ln t are the consequence of processes showing a broad and continuous distribution of relaxation times τ (or, similarly, a large hierarchy of energy barriers). This statement plus the analogy with experiments made in real spin glasses like CuMn [11] makes very plausible the existence of a superconducting glass state in this material mainly in a temperature range close to T_c. The conventional picture of flux creep with a single pinning barrier [9, 14] is certainly not sufficient to explain our observation. It might be a limit of the glass state at low temperatures only.

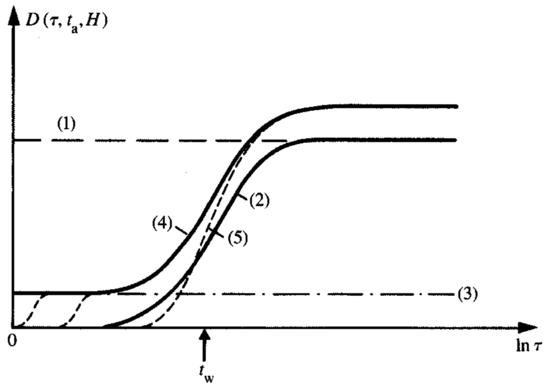

Figure 6

Schematic time evolution of the density of relaxation processes $D(\tau, t_a, H)$ for the superconducting glass cooled in H_0. During time t_w, $D_1(\tau, 0, H_0)$ (1) changes to $D_1(\tau, t_w, H_0)$ (2). The effect of ΔH at $t_a = t_w$ is the creation of second distribution $D_2(\tau, t_w, H_m = H_0 + \Delta H)$ (3), which is added to D_1 and yields $D = D_1 + D_2$ (4). As the short-time processes relax, D evolves toward $D(\tau, 2t_w, H_m)$ (5).

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