# Time-dependent magnetization of a superconducting glass

M. Tuominen and A. M. Goldman
*School of Physics and Astronomy, University of Minnesota, Minneapolis, Minnesota 55455*

M. L. Mecartney
*Department of Chemical Engineering and Materials Science, University of Minnesota, Minneapolis, Minnesota 55455*
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The time dependence of the magnetization of polycrystalline samples of the high-temperature superconductor $YBa_2Cu_3O_7$ has been investigated in order to ascertain the limits of the glasslike state. Slow rates of decay have been found to persist and to continuously decrease with increasing temperature up to the superconducting transition, suggesting that the latter is the upper bound on glasslike behavior.

The concept of glasslike behavior of superconductors was first discussed a number of years ago by Hertz,[1] who treated a continuum model with a random field. More recently, Stroud and co-workers[2] in a series of papers pointed out that the application of a magnetic field to a granular superconducting material would introduce frustration[3] into the Josephson coupling between grains,[4] and showed, using Monte Carlo techniques, that this would lead to behavior analogous to that of spin glasses. A randomly diluted lattice of Josephson junctions was found to be a glass in the analytical model given by John and Lubensky.[5] Glasslike properties were also found theoretically for regular two-dimensional (2D) Josephson-junction arrays, in certain magnetic fields.[6] Experimental evidence for the existence of a superconductive glass state was presented by Müller, Takashige, and Bednorz[7] (MTB), who inferred its existence from measurements of the susceptibility and magnetic moments of the high-$T_c$ superconductor La-Ba-Cu-O. Similar conclusions relating to the glasslike behavior of the same system were reached by Razavi *et al.*[8] and for $YBa_2Cu_3O_{7-\delta}$ by Carolan *et al.*[9]

In this paper we present the results of detailed measurements of the temperature dependence of the decay of the magnetization of bulk polycrystalline samples of the high-temperature superconductor $YBa_2Cu_3O_7$. We find many of the features of the so-called superconducting glass state reported by MTB, but the more extensive nature of our measurements of the decay of the magnetization leads us to somewhat different conclusions relating to the phase diagram. In particular, the locus of points denoting the intersection of the field-cooled and zero-field-cooled magnetizations which MTB have identified as the de Almeida–Thouless line[10] is not found to be a relevant boundary. A major feature of our measurements is that we are able to exclude conventional flux creep[11] as an explanation of the nonexponential time dependences of the magnetization over a substantial range of temperatures. Furthermore, if the glass phase is defined by the occurrence of long-time nonexponential decays of the magnetization, then we would conclude that the glass transition and the superconducting transition in the magnetic field are essentially indistinguishable. In contrast with conventional spin glasses where long decay times are a low-temperature property, the decay times appear to diverge as the superconducting transition is approached from below. The time dependence of the magnetic response above the superconducting transition temperature is that of a normal conductor.

Because these measurements have been carried out on polycrystalline aggregates, the behavior can be attributed at this time to the granular structure of the material and should not be considered to be an intrinsic property of the high-$T_c$ superconducting state, as has been suggested in a number of theoretical models.[12–14] However, preliminary measurements on single crystals which may have twin boundaries have yielded results very similar to those reported here, lending support to the view expressed in Ref. 7 that the characteristic size of the "grains" is nearly microscopic and smaller than the actual crystallites. Only an extension of these studies to defect-free single-crystal materials would establish the relevance of the present considerations to the intrinsic character of the superconducting state of high-$T_c$ materials.

The $YBa_2Cu_3O_7$ samples were prepared using standard solid-state reaction techniques[15] and determined to be the 1:2:3 phase by x-ray diffraction analysis. The existence of superconductivity was determined resistively using a four-point method. The onset of the transition was at 93 K and zero resistance was achieved at 90 K. The data reported in detail here were obtained using a sample which had a mass of 0.070 g, and dimensions of $1.1\times1.9\times9.0$ mm.[3] Scanning electron microscope analysis revealed a structure which consisted of elongated irregular plates with characteristic lengths of 10–20 $\mu$m and widths of 2–5 $\mu$m. Magnetic measurements were performed using a superconducting quantum interference device (SQUID) susceptometer.[16] All temperature values reported were determined using calibrated carbon-glass and platinum resistance thermometers provided with the susceptometer. The long axis of the sample was always aligned in the direction of the magnetic field, a configuration in which the demagnetizing factor was 0.05.

Field-cooled (FC) and zero-field-cooled (ZFC) magnetization measurements were made in fields ranging from 50 to 5000 G. Representative data is shown in Fig. 1 and is similar to behavior reported by MTB.[7] The data was

 

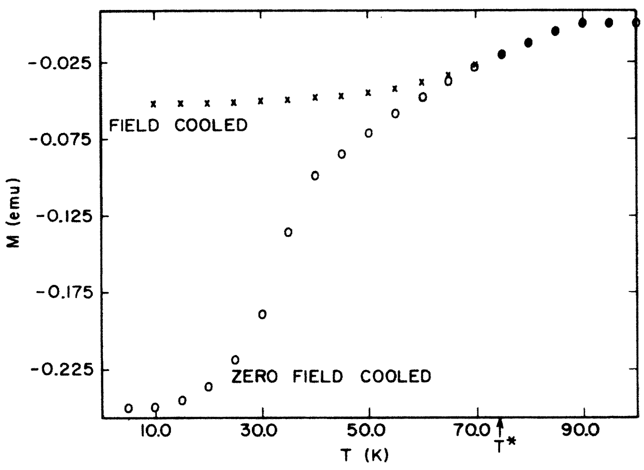


FIG. 1. Field-cooled and zero-field-cooled magnetization measurements of a $YBa_2Cu_3O_7$ sample taken using a field of 500 G.

obtained by first cooling the sample in zero field to 5 K, and then switching on a magnetic field and measuring the magnetization as a function of increasing temperature up through the value of $T_c$ for the material. Then the sample was cooled in a field and the magnetization was again measured while warming. A typical temperature sweep took three hours. The ZFC and FC curves intersect at a temperature MTB (Ref. 7) called $T^*$.

A difference between the ZFC and FC magnetizations is one of the salient features of a magnetic spin glass. Similar behavior would be expected of a phase glass or superconducting glass. Because the FC curves are reversible and the ZFC are not, $T^*$, the temperature at their intersection, has been identified by MTB (Ref. 7) as the temperature demarking the boundary between ergodic and nonergodic behavior. The boundary, or quasi-de Almeida-Thouless line,[16] was given by $T^*=T_c[1-(H/H_0)^{1/\gamma}]$. The present data are similar to that of (MTB) (Ref. 7) in this instance, with $H_0=7600$ G, $T_c=90$ $K$ and $\gamma\sim1.5$. However, as the ZFC and FC curves join tangentially, $T^*$ is really not well defined.

It should be noted that for a disordered and porous superconductor one might expect different magnetizations for the ZFC and FC cases simply because of different roles played by the Meissner effect and dc screening.[17] Consequently the proof of glasslike behavior would appear to depend upon the observation of nonexponential time decays in the magnetization when the external magnetic field is changed abruptly. Such decays were indeed observed by MTB,[7] and were cited as evidence for the glasslike character of the superconductivity. A difficulty with this is that there is another phenomenon associated with type-II superconductors, known as flux creep, in which trapped flux is also known to decay in a nonexponential fashion.[11]

Detailed studies of the dynamics have been carried out by measuring the decay of flux trapped in the sample as a function of time. This is in analogy to the thermoremnant magnetization in spin glasses. The sample was systemati-

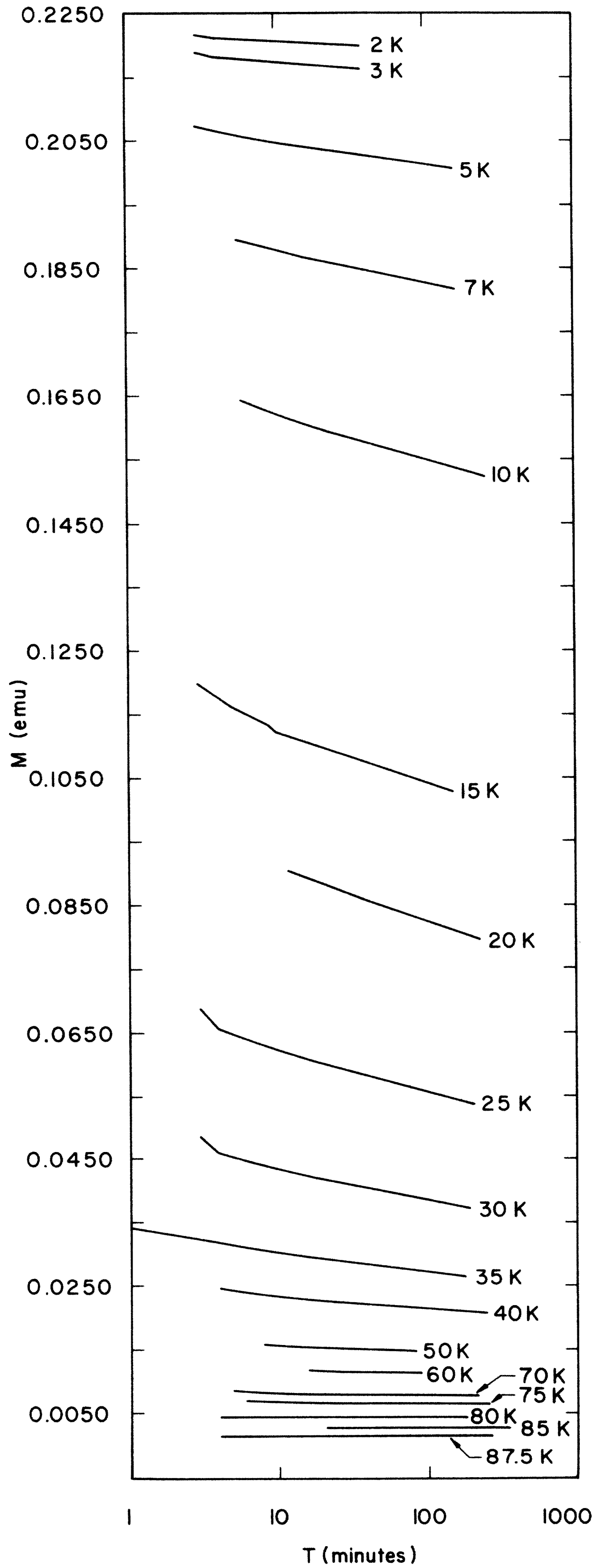


FIG. 2. Magnetization vs time of the sample studied in Fig. 1. Measurements were obtained by cooling in a field of 500 G and then removing the field. The residual magnetic field during these measurements was the order of 3 G.

cally cooled in a 500-G field to the temperature of interest, thereby introducing flux into the material. The applied field was then switched off. The magnetic response in the field quickly switched sign and the remaining trapped flux (paramagnetic signal) was found to decay linearly in $\ln(t)$ in a manner similar to remnant decay in spin glasses. Decays as a function of time at several temperatures are shown in Fig. 2.

Since the rate of decay is dependent on the magnitude of the trapped flux, each decay line should be normalized by dividing the rate by the magnitude of the initial trapped flux. In Fig. 3 we present the normalized rates of decay. $M_0$ corresponds to the flux trapped at $t=1$ min. It should be noted that the normalized decay rate continues to decrease as $T_c$ is approached from below and there is no discernible change going through $T^*$. These results suggest that the transition to glasslike behavior actually begins at $T_c(H)$ and not at $T^*$.[5] The measurements actually raise serious questions as to whether an equilibrium phase diagram of the type implied by Fig. 1 has meaning unless measurements are carried out over extremely long times.

It is important to note that there is a peak in the temperature dependence of the decay rate to about 30 K. Below this temperature the rate of decay increases with $T$; behavior consistent with both flux creep and the decay of the magnetization in real spin glasses. The observed continuing decrease of the rate with temperature above 30 K as the superconducting transition is approached from below suggests a kind of critical slowing down which, to our knowledge, is not contained in any model of either the spin glasses or the superconducting glass. The decay of the magnetization cannot be described as flux creep, the rate of which would increase with $T$.

Also suggestive of a glasslike state is our observation of the predicted instability of the ZFC curve.[2] Measuring the magnetization of points on this curve as a function of time, we have found them to decay linearly in $\ln(t)$ towards the FC curve, which is itself stable in time. The rate of this ZFC decay is also temperature dependent and

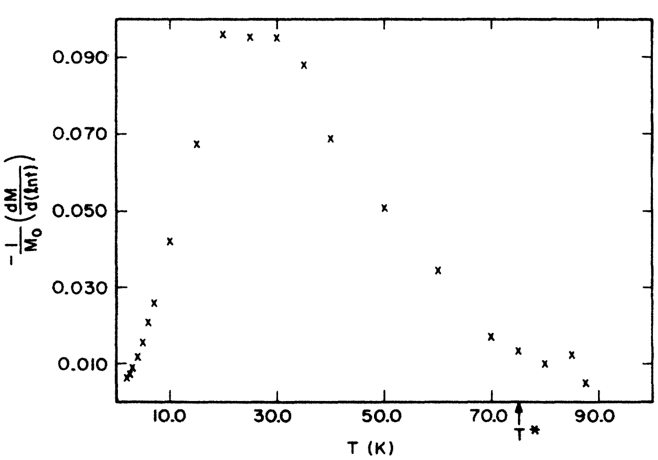


FIG. 3. Temperature dependence of the fractional change of the magnetization. $M_0$ is the magnetization measured 1 min after the removal of the external magnetic field.

has a maximum at 30 K in a field of 500 G. As the field is increased there is a decrease in the temperature at which the maximum is found. For the particular sample reported here, it shifts down to 5 K when the field is increased to 5000 G.

It should be noted that results similar to those described above have also been found for samples with different densities and processing histories and, as mentioned above, even in single crystals. The behavior is the same from sample to sample although the details of the transition temperatures and temperatures of the maximum in the decay rate did vary somewhat.

Although there are many formal similarities between spin glasses and the superconducting glass, it is important to realize in interpreting these data that there are differences which greatly weaken the analogy, as the measured magnetizations are related to the underlying order parameters in rather different ways. In a spin glass, the magnetization is directly related to the Edwards-Anderson[18] order parameter, whereas in the superconducting case it depends on macroscopic circulating currents which in turn depend on spatial gradients of the phase of the local order parameter. Furthermore, the magnetic field is the thermodynamic conjugate field to the magnetization in magnetic systems in which the tendency is towards ferromagnetic ordering, whereas in the superconducting case, where the intergrain coupling is ferromagnetic and $X$-$Y$-like in character, the applied magnetic field is not the conjugate field.

In the case of the spin glass, the application of a field reduces the disorder in the system, tending to align the microscopic magnetic moments. However, in the superconducting case the field itself is the origin of the frustration in which some of the couplings are ferromagnetic, whereas others tend to align pseudospins at angles other than 0°.

In summary, we have investigated the temperature dependence of the decay of magnetization in a granular superconductor. Nonexponential decays with long time constants have been observed all the way up to the transition temperature. The decay rate is an increasing function of temperature at low temperatures up to about 30 K. Above this temperature the decay rate decreases with increasing temperature with rapid response being restored only above $T_c$. This is different from the recent results of Mota *et al.*,[19] who report only an increase in $\partial M/\partial \ln t$ with $T$ in Sr-La-Cu-O and Ba-La-Cu-O, both of which are superconductors with lower values of $T_c$. Our results are suggestive of a superconducting glass which is rather different from the simplest analogies with spin glasses. In particular, the phase diagram suggested by MBT would appear to be relevant only on short time scales.

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