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Low-field microwave absorption in the superconducting copper oxides

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A nonresonant microwave absorption has been observed at fields below the thermodynamic critical field, $H_{c1}^{(T)}$, in the new copper oxide superconductors. This is associated with flux slippage and allows an estimation of the average area of the uniform phase in the superconducting glass state.

The recently discovered mixed copper oxide superconductors[1] have been shown to exhibit some properties of a superconducting glass.[2] The grains in these ceramics are not uniform bulk materials throughout but rather a composition of coupled superconducting clusters. A model for such a superconducting glass consisting of many weakly coupled clusters has been investigated using the Hamiltonian[3-5]

$$\mathcal{H} = -\sum_{ij} J_{ij} \cos(\phi_i - \phi_j - A_{ij}) \ .$$

The phase factors $A_{ij} = \kappa_{ij} H$ introduce randomness and frustration in the presence of a magnetic field H because the system has many competing ground states of almost the same energy. κ_{ij} is a random geometric factor. Hence, after cooling in zero magnetic field the system is in a Meissner or XY state.[3-5] On the application of a magnetic field, the system is weakly random and changes its configuration with accompanying flux slips starting at a critical field H_{c1}^{*}. The resulting magnetization of this state differs from that of a type-II superconductor in that it decays quite slowly after removing the field as some flux is trapped.[2,6] In granular metallic superconductors an Abrikosov vortex phase exists above $H_{c1}^{(T)}$,[4] and at still higher fields a transition occurs to a spin-glass-like state[3,5] with one quantum flux per loop.[4] We report here that the low-field microwave susceptibility $\chi''(H)$ below $H_{c1}^{(T)}$ is also determined by the glassy nature of the superconductivity in several of the new copper oxide-based superconductors[1,2,7,8] below T_c. Furthermore, an estimate of the area of the superconducting clusters may be obtained from $\chi''(H)$, showing they are smaller than or comparable to the ceramic particle size.

The dc magnetic properties of these new superconductors have been widely studied, and values of the lower critical field $H_{c1}^{(T)}$ are typically hundreds of gauss at low temperatures.[9] We have measured the high-frequency perpendicular susceptibility at magnetic fields below $H_{c1}^{(T)}$ in a Bruker or Varian 9-GHz ESR spectrometer. Various samples of the Ba-La, Sr-La, and Y-Ba copper oxides were investigated as listed in Table I. A small bar of the compressed sintered material of each compound with dimensions $3.5 \times 0.5 \times 0.5$ mm^3 was mounted in an Oxford Instruments continuous flow He cryostat with the externally applied magnetic field parallel to the long axis, thus making the demagnetizing field negligible. The sample in the cryostat is at the center of a microwave cavity so that the high-frequency magnetic field is perpendicular to the applied field, i.e., standard ESR geometry. The measured signal is the field derivative of the imaginary part of the perpendicular magnetic susceptibility as a function of the external magnetic field. Measurements were made for both increasing and decreasing field. Prior to each measurement, the sample was heated to about 50 K above its transition temperature and cooled in zero magnetic field (< 0.2 G) in order to attain a reproducible virgin state before each run.

Shown in Fig. 1 is the behavior of the $Ba_{0.15}La_{1.85}CuO_4$ sample for three different field sweeps of 50, 100, and 500 G. For each increasing field sweep, there is a maximum in $d\chi''/dH$ at ~ 13.5 G which is nearly reversible in the 50-G

TABLE I. Variation of H_{c1}^{*} in the different superconducting copper oxides used in the low-field microwave absorption experiments.

	H_{c1}^{*} (gauss)	Cluster radius mμ	T_c (K)
$Ba_{0.15}La_{1.85}CuO_4$	13.5	0.7	26
$Sr_{0.2}La_{2.8}Cu_2O_7$	0.8	2	35
$Sr_{0.2}La_{1.8}CuO_4$	0.6	2.3	40
$Y_{1.8}Ba_{1.2}Cu_3O_{7-\delta}$	0.5	2.5	90
$YBa_2Cu_3O_{7-\delta}$	~ 3–20	1.0–0.4	92
$YBa_3Cu_4O_{9-\delta}$	~ 8	0.6	92

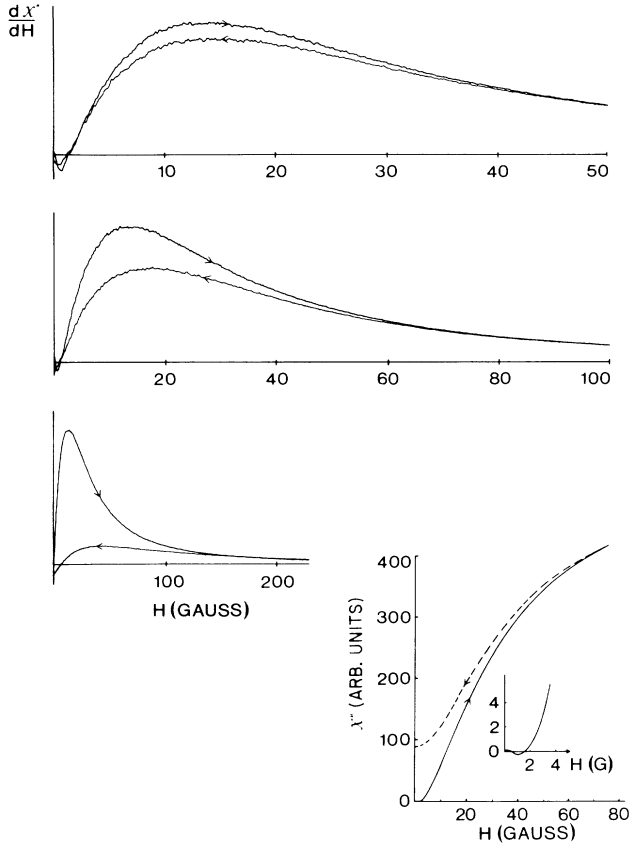

FIG. 1. Low-field microwave absorption in various field sweeps for $Ba_{0.15}La_{1.85}CuO_4$ at 4 K after zero-field cooling. The inset shows the integration of the 100-G field sweep.

decreasing sweep, but less so in the larger decreasing field sweeps of 100 and 500 G. The inset in Fig. 1 shows the integration of the 100-G sweep illustrating the field dependence of the lossy part of the microwave susceptibility χ''. It is apparent that the signal maximum corresponds to where the absorption changes most rapidly with increasing field and is associated with H_{c1}^* where flux slips start to occur.[3]

On decreasing the field back to zero, some of the flux becomes trapped, causing the weakly random state to persist partially at low fields. Hence, the microwaves can still penetrate the sample, giving an appreciable absorption signal at zero field. The amount of trapped flux depends on the extent of the magnetic field sweep, with more being trapped after a larger field sweep, thus inducing a larger remanent absorption. The magnetic properties of the sintered ceramic are clearly nonreversible for the first field sweep, but subsequent field sweeps nearly follow the curve obtained in decreasing field. Compared to the first field sweep after zero-field cooling, the signal maximum is now located at higher magnetic fields. A similar shift of the maximum is observed if the sample is cooled in an applied field. These nonreversible properties are typical of a superconducting glass.

Using the signal maximum as a measure of H_{c1}^* allows an estimation of an average projected area S of the superconducting loops in the glassy state:[3]

$$S = \phi_0 / 2H_{c1}^* ,$$

where ϕ_0 is an elementary flux quantum of 2×10^{-1} G cm^2. For $H_{c1}^* = 13.5$ G the loop radius is 0.7 μ.

The results for the strontium lanthanum copper oxides are all similar to those shown in Fig. 2 for $Sr_{0.2}La_{2.8}Cu_2O_7$.[10] The signal maximum occurs at a much lower field, $\sim$0.8 G, and the variation of the magnetization due to flux slips is clearly seen as a fluctuating signal when the field increases. The fluctuations in Fig. 2 occur closer together in larger field sweeps and therefore are not due to a noisy signal. This observation is further evidence that H_{c1}^* is near the signal maximum. The calculated superconducting loop radius in this case is 2 mμ. Also shown in Fig. 2 is the evolution of this new microwave absorption with temperature. It is clear that the absorption only appears in the superconducting state below T_c and is strongest at the lowest temperature measured.

The results differ among the various Y-Ba copper oxides. The sample with less barium than yttrium showed a signal maximum at the lowest field of all the samples measured, namely 0.5 G, but those with the approximate composition $YBa_2Cu_3O_{7-\delta}$ and $YBa_3Cu_4O_{7-\delta}$ had their maxima between 20 and 3 G. This means that while in the former sample the cluster radius is $\sim$2.5 mμ, the latter samples have cluster radii between 0.4 and 1.0 mμ. Two of the $YBa_2Cu_3O_{7-\delta}$ were annealed in oxygen which caused the superconducting transition to sharpen. The peak of the low-field microwave absorption was observed to shift from 20 to 16 G in one sample and from 16 to 3 G in the other, indicating that an increase in the area of the uniform phase also occurs on annealing. This illustrates how the low-field microwave absorption is a useful way to characterize these materials.

The signals observed in the Y-Ba copper oxides all showed larger fluctuations than those in Figs. 1 and 2. Some of these fluctuations correspond to structure in the signal because they are reproducible with increasing and decreasing fields. In one sample regular oscillations with a $\sim$120-G period were observed over the 10-kG range for a limited temperature range $\sim$8–12 K.

The average uniform phase size determined here ranges from 0.8 to 5 mμ in diameter. The particle sizes observed by transmission electron microscopy[11] for SrLaCuO, for example, vary by more than this, which means that while the smaller particles may be single phase most of the larger particles are composites of superconducting clusters. In the early samples which showed broad percolative transition behavior[1,2] the area of the uniform phase was considerably smaller, typically 0.03 mμ.[2] Similar samples of Estève *et al.*[12] showed the existence of Josephson junction inside the grains and determined from the Shapiro steps behavior under microwave irradiation $S = 0.1$ mμ.[2]

Some of the experiments presented here have been repeated at 19.3 GHz. The field-dependent microwave absorption resembles that found for 9.4 GHz, shown in Figs. 1 and 2, although no absolute comparison of χ'' was made. This consistency further supports our interpretation of the

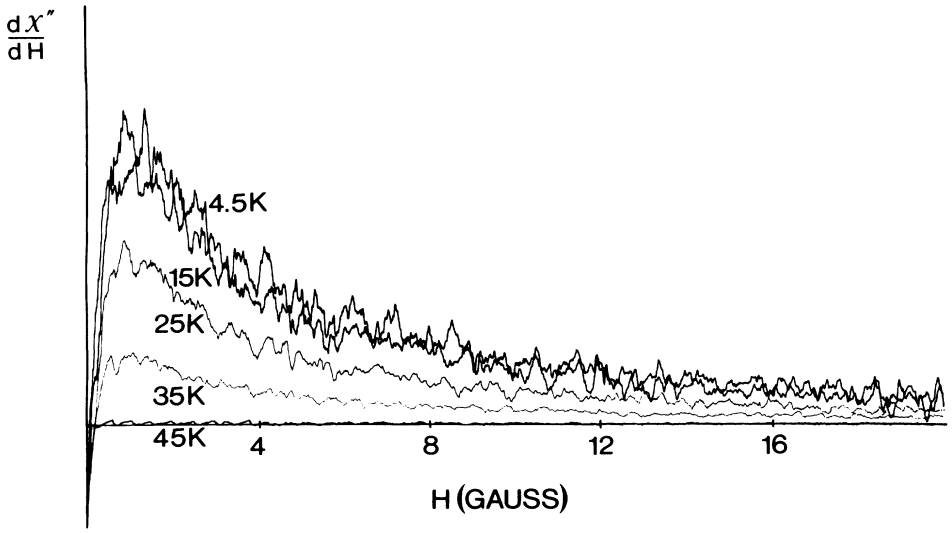

FIG. 2. Low-field microwave absorption of $Sr_{0.2}La_{2.8}Cu_2O_7$ at various temperatures after zero-field cooling.

cause of the microwave absorption.

In conclusion, the results of the low-field, nonresonant microwave absorption experiments are consistent with the static simulations[3-5] and magnetic properties of the $Ba_{0.15}La_{1.85}CuO_4$ compound[2] as well as the recently measured time decay of the magnetization, $M(t)$, at low temperatures and fields.[6,13] We have shown that all the new high-T_c oxide superconductors show glassy behavior in their ceramic form. Furthermore, the measurements presented here can be used to characterize the material in a simple way.

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