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Microwave absorption studies of Y-Ba-Cu-O

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We have measured the magnetic field dependence of the microwave absorption of bulk ceramic $Y_1Ba_2Cu_3O_x$ as a function of temperature. Below the superconducting transition temperature, the microwave absorption increases with increasing magnetic-flux penetration into the sample, and the absorption exhibits considerable hysteresis resulting from hysteresis in sample magnetization. Also, when the applied magnetic field is removed there is a remanent absorption due to the remanent sample magnetization.

It is well known[1] that superconductors absorb microwaves when $h\nu \gtrsim 2\Delta$, where ν is the microwave frequency, and 2Δ is the superconducting energy gap. Such microwave absorption results in quasiparticle creation, and the observed absorption threshold can be used to determine the size of the gap. But absorption can also occur when $h\nu \ll 2\Delta$ if the temperature T and/or the magnetic flux B penetrating the sample is nonzero. In this case, very low energy excitations are possible due to the presence of thermally excited quasiparticles, or due to pair breaking by the magnetic flux at the surface of the sample or in the core of a flux vortex, In the framework of the two-fluid model, this can be thought of as absorption by the normal (dissipative) fluid, where the fraction of normal electrons is given by $n_n = 1 - |\psi|^2$, and ψ is the superconducting order parameter. Since regions of high-B penetration are regions with low $|\psi|^2$, n_n will increase monotonically with increasing magnetic flux penetration, as will the microwave absorption. Measurements of microwave absorption as a function of applied magnetic field are thus a convenient contactless probe of field penetration, and thus a means of determining H_{c1} and H_{c2} of a type-II superconductor. It should be noted that because of coherence effects, Bardeen-Cooper-Schrieffer[1,2] (BCS) theory actually predicts that electromagnetic absorption in the limit $h\nu \ll 2\Delta$ will be greater in the superconducting state just below T_c than it is in the normal state. Such an increase will not be seen, however, if the volume fraction of the material sampled by the microwaves decreases fast enough due to a decrease in skin depth below T_c

We have made microwave absorption measurements on a variety of high-T_c superconductors and report here our results on bulk single-phase ceramic $Y_1Ba_2Cu_3O_x$. In addition to changes in absorption at H_{c1} and H_{c2}, we observed a large hysteresis in absorption, which we believe is due to hysteresis in sample magnetization. We also observed a large remanent absorption which exhibits the properties seen in magnetization measurements which have been attributed[3] to a superconductive glass phase.

Samples were cut from nominally single-phase $Y_1Ba_2Cu_3O_x$ with a superconducting-transition-temperature midpoint $T_c = 91.7$ K and a resistive transition width of 2 K. The superconducting material was prepared by grinding mixtures of the appropriate molar ratios of BaO (100 mesh), Y_2O_3 (325 mesh), and CuO (200 mesh). The ground mixture was pressed into disks at a pressure of 413 MPa (60000 psi). The disks were fired in air by ramping from 25 to 950 °C (3 h), sintering at 950 °C (12 h), and cooling in the oven to 25 °C (3.5 h). Further details of sample preparation can be found in Ref. 4.

The sample was placed in a resonant microwave cavity in the variable temperature cryostat of an Oxford Instruments superconducting magnet. The microwave source was a 100 mW Gunn oscillator tunable in the range 22 to 23 GHz. The microwave power reflected from the cavity was measured as a function of the applied magnetic field (H) and T. Other reports[5] of the field dependence of microwave absorption have all been based on measurements using magnetic field modulation. The field modulation technique is routinely used in EPR spectrometers to record the field derivative of the microwave absorption signal. It is important to note that in this instance, due to the presence of hysteresis in the microwave absorption, field modulation does not yield the derivative of the absorption. For this reason, we have not used field modulation in this study.

Above T_c we observed a large microwave absorption which was nearly independent of H. For T less than but near T_c the microwave absorption increased very dramatically as H was ramped through the resistive transition, and leveled off above H_{c2}, i.e., the microwave losses are greatest in the normal state. (This property allows one to easily measure H_{c2} as a function of T.) As the temperature was decreased further, H_{c2} increased very rapidly and soon exceeded the maximum field of our magnet (6 T). Also, for $T < T_c$, we observed a pronounced bend in the absorption versus H curve at very low H. This is illustrated in Fig. 1(a), for $T = 85$ K. At that T, we saw no hysteresis, i.e., the increasing (solid line) and decreasing (dashed line) field sweeps coincide. We believe the bend in the curve marks the position of H_{c1}. The reason for this assignment can be understood in the following way. In a ceramic superconductor, magnetic flux can penetrate between the grains when the intergranular Josephson screening currents begin to exceed the critical Josephson supercurrent.[6] The field at which this occurs depends on the magnitude of the Josephson coupling between grains and can be extremely small, even less than 1 G.[6] Although flux penetrates between the grains, it will penetrate only the surfaces of the individual crystallites until $H > H_{c1}$, at

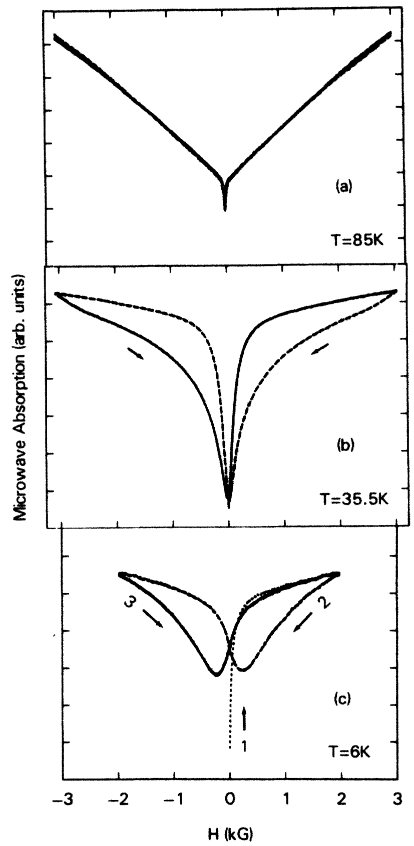

FIG. 1. Microwave absorption of granular Y-Ba-Cu-O vs applied dc magnetic field. (a) $T=85$ K. Solid curve $dH/dt>0$ and dashed curve $dH/dt<0$ coincide. (b) $T=35.5$ K. Solid curve $dH/dt>0$ and dashed curve $dH/dt<0$ form hysteresis loop. (c) $T=6$ K. Dotted curve, initial field application, $dH/dT>0$. Dashed curve, $dH/dt<0$, and solid curve, $dH/dt>0$, form subsequent hysteresis loop exhibiting large remanent absorption at $H=0$.

which point vortices will enter the individual grains. Since the surfaces of the crystallites are the region most accessible to the microwaves, the most rapid change in microwave absorption should occur below H_{c1}. Above H_{c1}, the surface penetration has saturated, vortices of flux penetrate the crystallites, and the cores of the vortices will be sampled by the microwaves a skin depth in from the surface. Such a crossover from surface sampling to core sampling could account for the decrease in slope which we observed above H_{c1}. If a quantitative value for H_{c1} is desired, the position of the bend in absorption must be corrected for demagnetization effects[1] Although this was not done in the present case, due to complicated sample geometry, the uncorrected value of approximately 100 G at 6 K is not unreasonable when compared to H_{c1}'s of several hundred gauss in single crystals.

Hysteresis in microwave absorption first appeared between 80 and 85 K. Since the absorption is related to the B flux in the sample, and $\mathbf{B}=\mathbf{H}+4\pi\mathbf{M}$, the observed hysteresis can be viewed as a natural consequence of hysteresis in sample magnetization, M. The area of the hysteresis loop increased as T was lowered, as did the value of H_{c1}. Figure 1(b) shows the large loop obtained at $T=35.5$ K. This loop has the same birdlike shape reported by Yan Shousheng *et al.*[7] for the field dependence of the penetration depth, as measured by the frequency shift of a circuit resonating near 200 kHz. This similarity supports the picture of microwave absorption tracking flux penetration.

Below 35 K, two local minima (symmetrically place about $H=0$) developed in the microwave absorption, and the birdlike shape was transformed into what looks more like a butterfly. The butterfly shape obtained at $T=6$ K appears in Fig. 1(c). The dotted line indicates the initial steep climb in microwave absorption when the field was first applied. This is followed by the downsweep (dashed) and upsweep (solid) which form the hysteresis loop. The position of each minimum moved away from $H=0$ as T was decreased and as the maximum applied field was increased. The local minima in microwave absorption probably result from a change in sign of the sample magnetization. Such a change in sign would allow the magnitude of B to go through a minimum at nonzero H. We attribute the observed remanent absorption (when the applied field was removed) to the remanent magnetization of the sample.

A narrower minimum is absorption was obtained by restricting H to positive values, as in Fig. 2. The dashed curve is the dependence on initial application of H, and the solid curve is the subsequent hysteresis loop. In the solid curve, there is an initial decrease in absorption on increasing the field from zero. We believe this decrease results from a cancellation of flux within the sample as the diamagnetic response of the sample has opposite sign from the remanent magnetization. According to this interpretation, the position of the minimum is correlated to the magnitude of the remanence, since a larger remanent magnetization requires a larger field for cancellation to occur. Figure 3 plots the position of the narrow minimum, at $T=5.2$ K, as a function of the maximum magnetic field

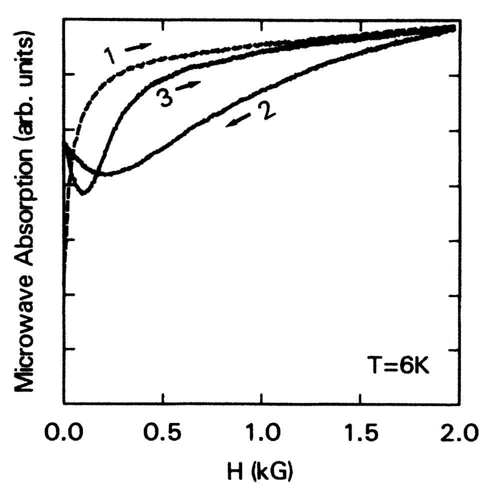

FIG. 2. Microwave absorption of granular Y-Ba-Cu-O vs applied magnetic field, $T=6$ K. Dashed curve, initial field application. Solid curve, subsequent hysteresis loop, H positive.

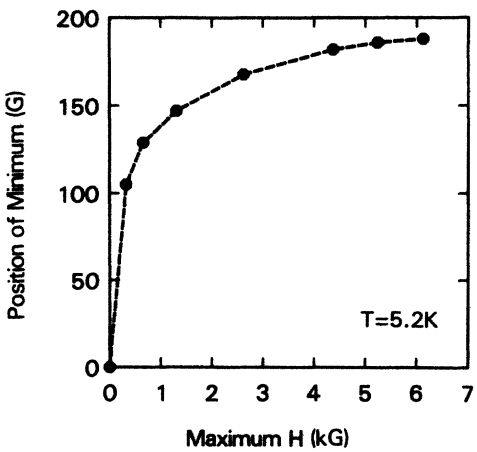

FIG. 3. Position of the narrow minimum in microwave absorption vs maximum applied magnetic field, reflecting the field dependence of the remanent magnetization at $T=5.2$ K.

applied. This figure illustrates the buildup of the remanent magnetization as the maximum applied field is increased. Most of the change occurs below 1 kG.

Because of the apparent correspondence between the microwave absorption signal and sample magnetization, we were able to observe, via microwave absorption, all of the features which have been attributed[3] to superconductive glass behavior—namely, the difference between field cooling and zero-field cooling, the boundary in H-T space separating ergodic from nonergodic regions, and logarithmic time decay of the remanence. These features were also observed by Yan Shoushen *et al.*[7] in their experiments at 200 kHz. They report an 85 K glass transition temperature for Y-Ba-Cu-O, which is approximately the same T below which we first observed hysteresis in the microwave absorption.

Nonresonant magnetic field-dependent microwave absorption in Y-Ba-Cu-O was first reported by Durny *et al.*,[5] although they do not include any data showing the field dependence. For $T < T_c$, they reported an absorption peak at zero field with a half-width of 20 G and a broad absorption which decreased slowly with increasing dc magnetic field. As described above, we observe an absorption minimum, not maximum, at zero field (except below 35 K where this minimum is shifted to nonzero field). Also, we see a broad absorption which increases, not decreases, with increasing field. In an absorption experiment, the microwave power reflected from the cavity can either decrease or increase, depending on whether the cavity is undercoupled or overcoupled. The sense of the resonant spin absorption can be used to distinguish between these two cases. In our experiment, relative to the resonant spin absorption, measured in the same sample at the same temperature, the zero-field absorption corresponds to an absorption minimum, not maximum. Durney *et al.*[5] attribute the nonresonant microwave absorption to an enhanced nonequilibrium ac diamagnetic susceptibility in the granular superconductor. We do not believe that the nonresonant absorption is a magnetic susceptibility absorption, but rather, that is is dominated by resistive losses, i.e., charge carriers absorbing energy when accelerated by the microwave electric field. Our conclusion is based on the observation that when the sample is located in a region of maximum rf magnetic field and minimum rf electric field, the spin resonance absorption (magnetic dipole transition) is maximized and the nonresonant absorption is minimized. Similarly, when the sample is situated such that the spin signal is minimized, the nonresonant absorption is maximized. Furthermore, we find that the absorption is maximum (although H independent) in the normal state, consistent with resistive losses. However, we cannot rule out the possibility of a small magnetic contribution to the nonresonant absorption signal.

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[1]For a general introduction to superconductivity, see M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, New York, 1975).

[2]J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. **108**, 1175 (1957).

[3]K. A. Müller, M. Takashige, and J. G. Bednorz, Phys. Rev. Lett. **58**, 1143 (1987).

[4]W. J. Gallagher, R. L. Sandstrom, T. R. Dinger, T. M. Shaw, and D. A. Chance, Solid State Commun. **63**, 147 (1987).

[5]Carlos Rettori, Dan Davidov, Igal Belaish, and Israel Felner, Phys. Rev. B **36**, 4028 (1987); R. Durny, J. Hautala, S. Ducharme, B. Lee, O. G. Symko, P. C. Taylor, D. J. Zheng, and J. A. Xu, *ibid.* **36**, 2361 (1987); S. V. Bhat, P. Ganguly, T. V. Ramkakrishnan, and C. N. R. Rao, J. Phys. C **20**, L559 (1987); K. W. Blazey, K. A. Müller, J. G. Bednorz, W. Berlinger, G. Amoretti, E. Buluggiu, A. Vera, and F. C. Matacotta, Phys. Rev. B **36**, 7241 (1987); K. Khachaturyan, E. R. Weber, P. Tejedor, Angelica M. Stacy, and A. M. Portis, *ibid.* **36**, 8309 (1987).

[6]John R. Clem and Vladimir G. Kogan, Jpn. J. Appl. Phys. **26**, Suppl. 26-3, 1161 (1987).

[7]Yan Shousheng, Jia Qiuping, Wang Xu, Xia Guoqiang, and Ma Hong, Progress in High-T_c Superconductivity, Vol. II (unpublished).