Microwave absorption of the high-T_c superconductor $YBa_2Cu_3O_7$

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Using a resonant-cavity perturbation technique, the microwave absorption at 16.8 GHz of the high-T_c compound $YBa_2Cu_3O_7$ is measured as a function of temperature (4–300 K) and magnetic field (0–7 T). At zero field the microwave loss shows a rapid decrease below 93 K, the onset of the superconducting transition. An important absorption, which is a decreasing function of temperature, is observed in the superconducting state. When a magnetic field is applied, the transition is broadened and shifted toward lower temperatures: an upper-critical field $H_{c2}(T=0) \sim 150$ T is obtained via extrapolation of the data. In the superconducting state, the absorption is found to be highly dependent on the magnetic-field intensity. For magnetic fields above 500 Oe, the absorption is a slowly increasing function and it is understood in terms of the temperature behavior of H_{c2}; for $H < 200$ Oe, the absorption increases very rapidly with field and it is believed to be correlated to H_{c1} and to the anisotropic character of the structure.

I. INTRODUCTION

The discovery of superconductivity above 90 K in the Y-Ba-Cu-O compounds[1] has initiated a lot of theoretical and experimental studies in order to understand the nature of the mechanism which triggers the superconducting transition. In order to pursue this task, magnetic and transport measurements[2] are systematically performed on these compounds. A dc resistivity measurement is generally concerned with the normal state above the superconducting transition while the magnetization deals with the superconducting state below the transition. The determination of the critical fields H_{c1} and H_{c2} are usually related to both measurements.

The ac techniques may also be of great interest to obtain information on the normal and superconducting states. Among these, far infrared spectroscopy has been widely used[3,4] on high-T_c compounds. This technique gives access to the real and imaginary parts of the complex ac conductivity and, thus, offers the possibility to determine an eventual energy gap. Information on the structural (phonons) and electrical (plasma frequency) properties can be obtained in this way. Microwave techniques which have also been used for the study of these compounds[5,6] should be very efficient to characterize the superconducting transition. We have recently used a microwave cavity perturbation technique to study the compound $La_{1.8}Sr_{0.2}CuO_4$.[7] The technique is rather well adapted to measure the resistivity in the normal state with high precision and to characterize the superconducting transition in an applied magnetic field. In the present paper, we report similar data obtained by the same technique on the compound $YBa_2Cu_3O_7$. The superconducting transition is fully characterized in magnetic fields from 0 to 7 T and a limit is given for the upper-critical field $H_{c2}(T=0)$. From the finite microwave absorption below the transition, it is also possible to analyze qualitatively and quantitatively the behavior of H_{c1}. The versatility of the technique for high-T_c superconductor studies is underlined and discussed.

II. MICROWAVE ABSORPTION

Microwave absorption in normal metals and superconductors has been the object of several studies and experiments.[8,9] Such measurements are, indeed, considered very useful to get information about the superconducting state below the temperature where the dc resistance vanishes. Because the skin depth at microwave frequencies is very small in these materials, the measurements are usually done on very thin films whose quality must be controlled accurately. This has been done, for example, on superconducting granular aluminum films[10,11] and good agreement is generally obtained between theory and experiment.

With high-T_c superconductors, thin-film technology is still under developmment and we, therefore, have to rely on experiments with available bulk sintered samples. Fortunately, these compounds have a relatively low conductivity ($10^5\ \Omega^{-1}\ m^{-1}$) in the normal state which gives a rather large skin depth ($\sim 10\,\mu$m) of the order of the average grain size. In a microwave experiment we may then expect a substantial absorption which could be useful to characterize the superconducting transition. This has, indeed, been verified in $La_{1.8}Sr_{0.2}CuO_4$.[7]

For this microwave study we used a standard resonant-cavity perturbation technique.[12] A resonating cavity (TE_{102} mode) is perturbed by the insertion of a small conducting sample in its stationary microwave electric field. This results in a displacement of the resonance frequency f, and a variation of its quality factor Q. If the sample is approximated by an elongated ellipsoid, we may get the complex dielectric function $\epsilon = \epsilon_1 + j\sigma/\omega\epsilon_0$ when the field is uniformly distributed inside the sample (quasistatic approximation); $\epsilon_1, \omega, \sigma$,

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and ϵ_0 are, respectively the dielectric constant, the angular frequency, the conductivity, and the free space permittivity. This is simply done by measuring both the relative frequency shift $\Delta f/f$ and the quality factor variation $\Delta(1/2Q)$.

In metals and superconductors, the microwave electric field is not uniform in the sample and the quasistatic approximation is inadequate. For that case, the field penetrates only over a small distance at the surface and the skin depth approximation equations[13] are used, instead, to evaluate the conductivity

$$\Delta f/f = -\alpha/N, \quad (1)$$

$$\sigma = [\alpha 2b/N^2\Delta(\tfrac{1}{2}Q)]^2(81\pi^5 f^3\epsilon_0/2^8c^2), \quad (2)$$

where α, N, $2b$, and c are, respectively, the cavity filling factor, the depolarization factor (of the ellipsoid), the sample's diameter, and the speed of light. In that case, we do not have access to the dielectric constant and the relative frequency shift is a mere geometrical factor. By measuring the microwave loss $\Delta(\frac{1}{2}Q)$ as a function of temperature, the behavior of the conductivity in the normal state may be obtained. Absorption below the superconducting transition is more difficult to explain since it can be the result of many effects, one of them being the fraction of the sample volume which is not transforming, but, if we assume that all the sample is effectively superconducting below T_c, it is convenient to introduce, following Tinkham and Glover,[14] a complex conductivity $\sigma_s = \sigma_{s1} + j\sigma_{s2}$ for the superconducting state. Following the treatment of Mattis and Bardeen,[15] we may expect an absorption in the superconducting state and a displacement of the resonance frequency at the transition.

III. SAMPLE AND EXPERIMENT

Polycrystalline samples of $YBa_2Cu_3O_7$ were prepared from appropriate amounts of $BaCO_3$, Y_2O_3, and CuO (all of 99.999% purity), mixed together and heated in air in an alumina crucible. The sample was held at 950 °C for 5 days in air with multiple grindings. X-ray diffraction was carried out on the powder and the trace indicated randomly oriented grains and the presence of a single phase. The powder was pressed into pellets and first annealed for 2 h in an O_2 atmosphere at 930 °C and then for 3 h in O_2 at 363 °C. Resistivity and magnetic susceptibility measurements were performed on the pressed pellets. The sample showed superconductivity with a transition midpoint of 92 K and a transition width of 2.5 K: the room temperature dc resistivity value was approximately 2000 $\mu\Omega$ cm and the value just above the transition 1000 $\mu\Omega$ cm. In the magnetic susceptibility measurement, 52% shielding was obtained. This is the percentage generally observed for such sintered samples.

The sintered samples were cut from a pressed pellet and had typical dimensions $3\times0.4\times0.4$ mm^3, a geometry which may be approximated with sufficient accuracy by an elongated ellipsoid. The sample was glued on a quartz rod going through the cavity; it could then be easily inserted at the location of the maximum electric field of a cavity resonating in the TE_{102} mode at 16.8 GHz. As the microwave frequency is swept through the cavity resonance, the transmitted power curve is registered at each temperature, in order to measure the relative frequency shift and the quality factor variation.

The cavity and sample were placed in a double calorimeter head filled with helium exchange gas to ensure temperature stability and subsequently immersed in a liquid-helium bath. For magnetic-field measurements up to 7 T, the setup was placed in a Nb-Ti superconducting coil. The temperature was monitored and stabilized with a Lakeshore controller with either a Si diode sensor or a $SrTiO_3$ capacitor for the magnetic-field data. An overall precision of 0.2 K was obtained for the 4–300 K temperature range.

IV. RESULTS AND DISCUSSION

The measured relative frequency shift at 16.8 GHz is $\sim 1.12\times10^{-2}$ and it is practically temperature independent; this value is in agreement with the geometrical factor α/N. When the sample becomes superconducting, this value drops by only $\sim 2\times10^{-5}$: such a small effect at the transition has also been observed in the La-Sr-Cu-O compound ($\sim 4\times10^{-5}$)[7] and it, thus, seems to be characteristic of the high-T_c superconductors. Although this variation is real, it cannot be used in a reliable way because its reproducibility is limited by the stability of our rf sweeper. Therefore, the frequency shift data will not be discussed in this paper.

The microwave loss data $\Delta(1/2Q)$ at 16.8 GHz are presented in Fig. 1(a) for $4 < T < 300$ K. For $T > 93$ K, the loss increases almost linearly with temperature. Below 93 K it decreases first rapidly by $\frac{2}{3}$ at 90 K, then more slowly down to the lowest temperatures around 4 K. This rapid decrease is

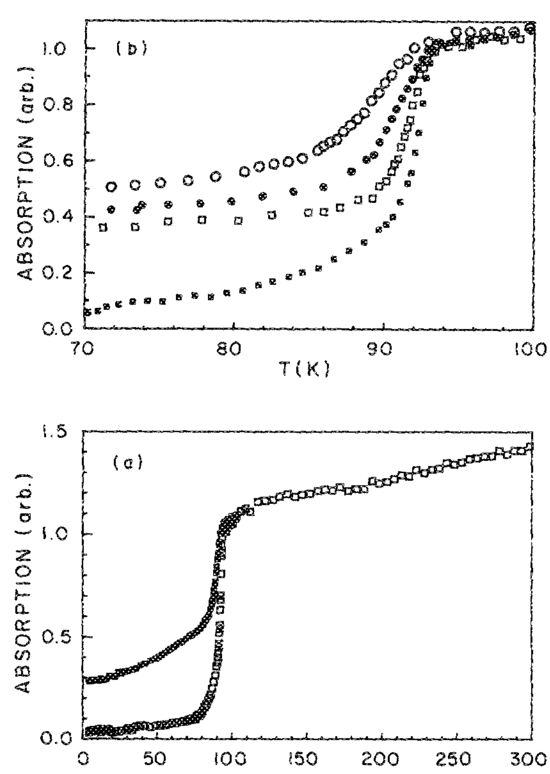

FIG. 1. (a) Microwave absorption at 16.8 GHz as a function of temperature: (□) 0; (■) 7 T. (b) Microwave absorption at 16.8 GHz as a function of temperature: (■) 0; (□) 1; (●) 3; (○) 7 T.

consistent with the superconducting properties observed through resistivity and susceptibility measurements. If these data are used in Eq. (2), one gets 180 $\mu\Omega$ cm for the room-temperature value of the resistivity and 100 $\mu\Omega$ cm just before the transition with a quasilinear decrease with temperature. These values are smaller than the one obtained by the dc technique. This is, however, consistent with the nature of the sintered sample which should offer more resistance to a direct current. The onset of the transition in our sample is at 93 K with a midpoint at 92 K: the width is difficult to evaluate because of a slowly decreasing loss below 90 K. The loss observed below the transition is quite important and it is not believed to be the result of a fraction of the sample which is not transforming as the x-ray diffraction analysis indicated the presence of the single phase. This absorption below T_c will be discussed shortly.

When a magnetic field is applied perpendicular to the sample axis, the loss data are modified only for $T < T_c$: these are presented in Fig. 1(b) for $70 < T < 100$ K as a function of different magnetic-field intensities. As it is easily seen in the figure, the microwave loss below the transition increases rapidly for small fields ($H < 1$ T), then more slowly as the field reaches 7 T. The observed magnetic-field dependence of the transition is typical of what is generally found in the superconducting state.[1] The rapid decrease below 93 K is shifted toward lower temperatures with increasing magnetic field, while the width of the transition is increasing. According to these data, a linear relation is found between the applied field and the midpoint transition temperature (midpoint taken relative to the plateau observed in the loss around 70 K); by extrapolation, a fairly large upper-critical field $H_{c2}(T=0) \sim 150$ T is deduced with a temperature dependence $-dH_{c2}/dT|_{T_c} = 2.36T/K$. These values are consistent with the bounds established on samples of similar composition.[16]

In Fig. 2 we present the normalized loss data A_s/A_n as a function of the reduced temperature T/T_c for 0 and 7 T magnetic field: $A_n = \Delta(1/2Q)_n$ is the loss evaluated just before the transition and $A_s = \Delta(1/2Q)_s$ the loss in the superconducting state. In the zero magnetic field the observed behavior is similar to the data obtained by others[5] on samples of comparable composition in the same frequency range. Qualitative agreement with the behavior generally observed on conventional superconductors is obtained. For this last case the Mattis–Bardeen expressions[15] for the conductivities in the local limit are used together with the BCS coherence factors and density of states to get the surface impedance of an impure superconductor. According to this approach,[5] the temperature dependence of A_s/A_n arises solely from the gap $\Delta(T)$. In our case, many parameters are unknown (penetration depth, coherence length, skin depth) and the data fitting will necessitate too many adjustable parameters, which is why we will not pursue, here, a quantitative analysis. We must, however, point out the potential usefulness of the technique to deduce the temperature dependence of the gap if the BCS theory is used. Contrary to what has been measured by others,[5] we did not observe any detectable structure in our data: the temperature range covered here is actually much larger than the previous reports[5] since we measure a stronger absorption.

If the same model is retained for the $H = 7$ T curve in Fig. 2, the dramatic increase of the loss relative to the zero field should be related to $\Delta(H)$. To test this hypothesis we have examined the field dependence in more details. The loss data obtained at very low magnetic-field intensities are presented in Fig. 3 for three selected temperatures below T_c. As it can be seen, the loss increases sharply in a quasilinear way for small magnetic fields below a characteristic value H_m, the value of H_m being itself a decreasing function of temperature; at 4 K, the value of H_m is around 150 Oe. For $H > H_m$ the loss increases more slowly with a slope increasing with temperature. This second regime is more apparent in Fig. 4 where the range of the magnetic field is larger.

According to data presented in Figs. 3 and 4, we distin-

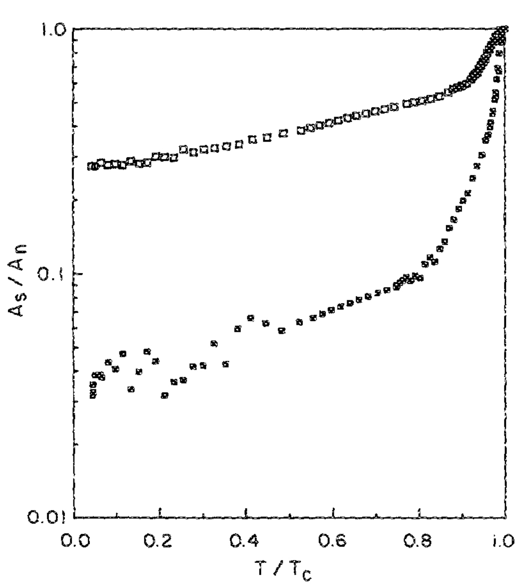

FIG. 2. Normalized microwave loss A_s/A_n as a function of the reduced temperature T/T_c: (■) 0; (□) 7 T.

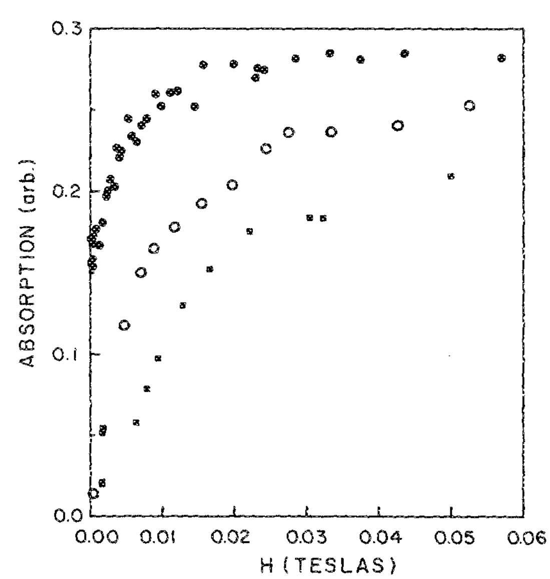

FIG. 3. Microwave loss at 16.8 GHz as a function of the magnetic field: (■) 4.2; (○) 40; (●) 80 K.

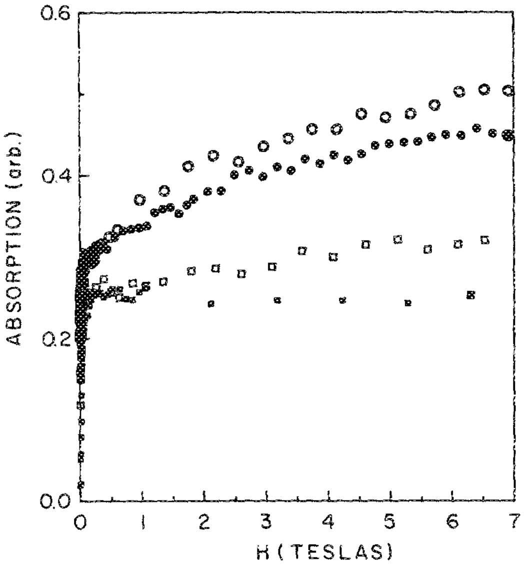

FIG. 4. Microwave loss at 16.8 GHz as a function of the magnetic field: (■) 4.2; (□) 40; (●), 70; (○) 80 K.

guish clearly two regimes of magnetic-field dependence for the microwave loss in the superconducting state. At low fields ($H<200$ Oe) it is very tempting to associate the observed behavior to the lower-critical field H_{c1} as the values of H_m are of the same order of magnitude as the ones reported in the literature[17]: H_m decreases with temperature as H_{c1} also does. At high fields the magnetic-flux lines penetrate the sample progressively with a vortex structure and the microwave loss is seen to increase; this increase with the field is more pronounced at high temperatures since the critical field H_{c2} is generally a decreasing function of temperature.

For $H_{c1} < H \ll H_{c2}$ the data are likely to be explained by the vortex structure which allows the coexistence of superconducting and normal regions. The increase of the microwave absorption is then due to increasing numbers of vortices. The dissipation can come from the normal core of the vortices.[18] In the absence of flux pinning, the relative loss in conventional type II superconductors[18] is expected to be proportional to the ratio H/H_{c2}. As we measured here, at 7 T, an absorption which is around 1/3 of the full normal value, we should have expected $H_{c2} \sim 20$ T, a value much smaller than the 150 T obtained from another portion of the curve. This difference may be a reflection of a large anisotropy ratio in this superconductor. At low fields ($H \ll H_m$) the rapid increase of absorption is not yet understood, but it could be related to the anisotropic nature of the Y-Ba-Cu-O compounds. Recent results[19] have indicated an anisotropy ratio $H^{\parallel}_{c2}/H^{\perp}_{c2} \sim 25$–$50$, where $H^{\parallel}_{c2}$ is for the field, parallel to the copper-oxygen layers and $H^{\perp}_{c2}$ for the perpendicular orientation. The expected anisotropy[10,20] for H_{c1} is $H^{\parallel}_{c1}/H^{\perp}_{c1} \lesssim H^{\perp}_{c2}/H^{\parallel}_{c2}$. As the sample is polycrystalline, the microwave measurement constitutes an average of the absorption along both directions. The application of a small magnetic field (~ 10 Oe) may be sufficient to allow vortex penetration along the parallel direction. At low fields, we should then expect a rapid increase of the absorption (Fig. 3). Above $H^{\perp}_{c1}$ (>200 Oe), vortices also start to penetrate the planes and the increase will be slower considering that the applied field is much lower than both $H^{\parallel}_{c2}$ and $H^{\perp}_{c2}$.[21] This is again consistent with the experimental data. Worthington, Gallagher, and Dinger[21] have determined a $H^{\perp}_{c1} \sim 5000$ Oe: this value is not consistent with the measured H_{c2} and predicted H_{c1} anisotropies. No structure at this field value is seen in our data and we believe that $H^{\perp}_{c1}$ is more likely to be around 200 Oe.

This anisotropy could also be reflected in an energy gap $\Delta(T)$ since we have observed a fairly large absorption in the zero magnetic field at low temperatures. In a surface impedance measurement Cohen *et al.*[22] have also observed a large absorption in the same temperature range and they explained their results in terms of superconducting grains coupled only weakly through insulating dielectric layers. We do not believe this issue of granularity to be adequate for our data: Cohen and co-workers measured an absorption, practically temperature independent, while our data show a well-defined temperature behavior in agreement with the expected variation of the superconducting properties. Anisotropy is, therefore, more likely to explain both temperature and magnetic-field behaviors.

V. CONCLUSION

In this paper, we analyzed in detail the temperature and magnetic-field dependencies of the microwave absorption of the high-T_c compound $YBa_2Cu_3O_7$. The temperature dependence of the superconducting transition is similar to what is measured in the dc resistivity. An important absorption was, however, observed below T_c. This could be attributed to thermally excited quasiparticles although the rather slow decrease with temperature indicates that a simple isotropic BCS gap is insufficient to account for the data. In a magnetic field, the transition is broadened and displaced toward lower temperatures. An upper-critical field $H_{c2}(T=0)$ of ~ 150 T was found by extrapolation. The residual absorption in the superconducting state is found to be highly modified by the magnetic field: two distinct field dependencies were observed. At high fields the increasing absorption was related to the normal regions induced by the vortex structure: at low fields the fast increase is not fully understood but a connection with H_{c1} is likely to be adequate if we consider the magnitude and temperature dependence of this field. The field behavior of such a polycrystalline sample is likely to be connected with the anisotropic nature of the compound, and in particular, that of the critical fields H_{c1} and H_{c2}. The large microwave loss observed at low temperatures and only moderate magnetic fields could constitute a significant problem for potential applications with these polycrystalline compounds.

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