PHYSICAL REVIEW B VOLUME 37, NUMBER 1 1 JANUARY 1988

Millimeter-wave absorption in La-Ba-Cu-O and Y-Ba-Cu-O superconductors

A. T. Wijeratne, G. L. Dunifer, J. T. Chen, and L. E. Wenger
Department of Physics, Wayne State University, Detroit, Michigan 48202

E. M. Logothetis
Research Staff, Ford Motor Company, Dearborn, Michigan 48121
(Received 14 September 1987)

Millimeter-wave absorption in the new high-T_c superconducting materials La-Ba-Cu-O and Y-Ba-Cu-O has been studied at a frequency of 80 GHz over the temperature range 4–300 K and in the presence of magnetic fields 0–5.5 T. It is found that there are strong correlations between the changes in microwave absorption and the superconducting transitions at 30 K for La-Ba-Cu-O and at 90 K for Y-Ba-Cu-O samples. The Y-Ba-Cu-O samples display additional microwave features near 250 K which may be associated with a higher-temperature superconductivity.

Observations of millimeter-wave absorption in the new high-T_c superconducting materials La-Ba-Cu oxide[1] and Y-Ba-Cu oxide[2] have been made at a frequency of 80 GHz over the temperature range 4-300 K and in the presence of magnetic fields 0-5.5 T. The absorption is monitored by measuring the propagation of microwave power around samples which mostly fill a section of waveguide. The propagation displays a sudden enhancement at temperatures below T_c. For the Y-Ba-Cu-oxide samples an enhancement is noted at both $T \approx 90$ K and $T \approx 250$ K. The effect of the magnetic field is to decrease the enhancement of propagation and to reduce the temperature at which it begins.

Three types of samples having nominal compositions of $La_{1.8}Ba_{0.2}CuO_{4-x}$, $YBa_2Cu_3O_{7-x}$, and $Y_5Ba_6Cu_{11}O_x$ were investigated. All were prepared by the solid-state reaction method with Table I giving the details of the preparation conditions. An appropriate mixture of the starting materials in powder form was first heated in air. The reacted mixture was then pulverized, pressed into pellets or bars, and sintered in air. X-ray powder diffraction patterns showed for the $YBa_2Cu_3O_{7-x}$ samples a dominant single phase with identical lines to those previously seen[3] while multiple phases were seen in the $Y_5Ba_6C_{11}O_x$ samples.

Figure 1 shows a schematic diagram of the spectrometer which was used. The superconducting specimen fills a section of waveguide of length L situated between a klystron, the microwave source, and a sensitive detector. The spectrometer is a modified version of the transmission spectrometer which has been used to study the propagation of microwaves through thin metal samples.[4] The power incident on the sample can be varied with an attenuator over a range of 50 dB up to a maximum of 100 mW. The detector is a liquid-helium-cooled InSb bolometer operating in a hot-electron mode. It can be used as a straight power detector (sensitivity of 10^{-14} W) or as a homodyne mixer (sensitivity of 10^{-21} W) with the use of a microwave reference of variable amplitude and phase obtained from the original klystron. A dc magnetic field aligned with the axis of the waveguide is obtained from a superconducting solenoid in which the sample is centered. The sample is situated in a vacuum can immersed in liquid helium. The use of a heater and thermometer allows its temperature to be varied continuously between 4 K and room temperature.

The internal dimensions of the rectangular waveguide are $a = 3.10$ mm and $b = 1.55$ mm. The oxide samples are cut into the form of rectangular bars with a low-speed diamond saw and then lapped on abrasive paper to dimensions about 0.04 mm smaller than the waveguide so that they slide easily into the waveguide interior. A typical length of sample is $L = 5.0$ mm. Our observations are independent of the incident power level except for some

TABLE I. Details of sample preparation and results from x-ray powder-diffraction patterns.

Nominal composition	$La_{1.8}Ba_{0.2}CuO_{4-x}$	$YBa_2Cu_3O_{7-x}$	$Y_5Ba_6Cu_{11}O_x$
Starting materials	La_2O_3, $BaCO_3$, and CuO	Y_2O_3, $BaCO_3$, and CuO	Y_2O_3, $BaCO_3$, and CuO
Reaction temperature (°C)	900	900	900
Reaction time (h)	8+pulverizing+6	8	8
Pressure for forming pellets (psi)	8000	8000	8000
Sintering temperature (°C)	1100	900	1100
Sintering time (h)	4	8	8
X-ray powder-diffraction patterns	Single phase with K_2NiF_4-type structure and trace (<5%) of $Cu_2La_2O_5$	Single phase with other phases <2%	Multiple phases containing Y_2BaCuO_5 (dominant) $YBa_2Cu_3O_{7-x}$, $Y_2Cu_2O_5$, Y_2O_3, and CuO

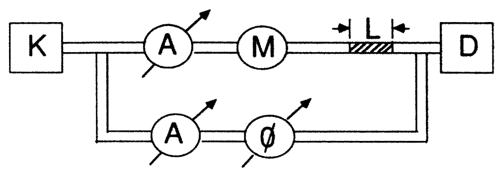

FIG. 1. Schematic representation of the spectrometer. The components include K, klystron; A, variable attenuator; ϕ, variable phase shifter; M, modulator; and D, detector.

heating of the sample at the highest levels. At 4 K a typical reflection coefficient from the sample is 3–5 dB, whereas a typical transmission coefficient for microwave propagation *around* the sample is 10–15 dB. Thus, the sample absorbs the majority of the incident power, even though a significant amount appears at the detector. The microwave power is clearly traveling around the sample and not through it, as has been demonstrated by replacing the sample with a solid Pb sample of similar dimensions and observing a comparable power level at the detector. Also, by using electrically conducting silver paint to fill the gap between the oxide sample and the waveguide, the power level at the detector drops 150 dB or more.

The microwaves propagate down the waveguide from the klystron in the TE_{10} mode with the rf electric field perpendicular to the broad faces of the waveguide. Because the cutoff frequency for this mode, $f_c = c/2a$, is independent of b, it appears that a significant quantity of microwave power continues to propagate in the TE_{10} mode in the narrow gaps between the superconducting sample and the broad faces of the silver waveguide. In fact, the power level at the detector displays certain characteristics of a "double slit" as the frequency is changed due to variation in the interference between the two waves that propagate in the top and bottom gaps. By sealing one of the gaps with silver paint, such variations are considerably reduced.

Figure 2 shows a semilog plot of the transmitted power as a function of the temperature for a $YBa_2Cu_3O_{7-x}$ sample as well as the dc electrical resistance of the sample. T_c (measured at the midpoint of the dc resistance step) is 90 K, and the width of the transition ΔT (measured between 10% and 90% of the resistance step) is 4 K. Figure 3 shows a similar plot for a $Y_5Ba_6Cu_{11}O_x$ sample having $T_c = 91$ K and $\Delta T = 4$ K. As can be seen for both samples, with decreasing temperature there is a sharp increase in the transmitted power occurring at T_c and having a width comparable to the ΔT measured from the electrical resistance. This feature is quite stable and remains even after a large number of thermal cyclings between 4 K and room temperature. For the $YBa_2Cu_3O_{7-x}$ sample there is a similar increase in the transmitted power occurring around $T \approx 265$ K even though there is no obvious step in the resistance curve. Although no step in the transmitted-power curve is observed in this vicinity for the $Y_5Ba_6Cu_{11}O_x$ sample, there is a distinct change in the slope at $T \sim 235$ K that coincides with a change of slope in the resistance curve. These higher-temperature features are not stable and degrade considerably after a few thermal cyclings. Lack of stronger correlations at these

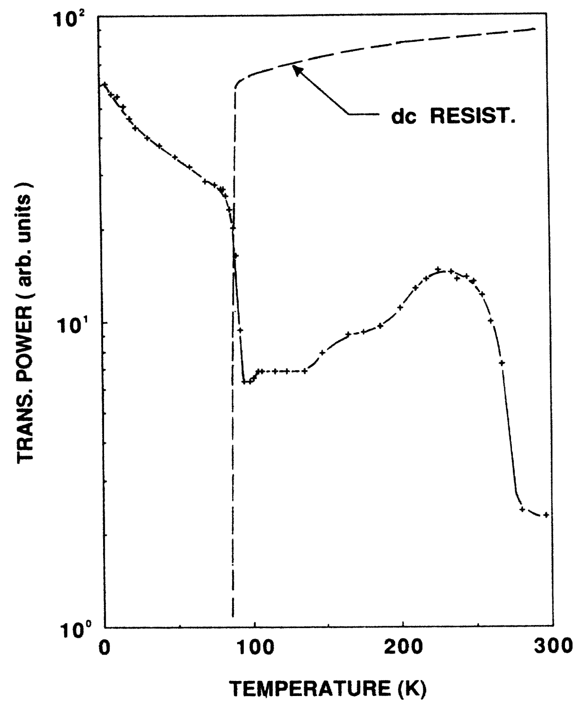

FIG. 2. Temperature dependence of the transmitted microwave power and dc electrical resistivity of a $YBa_2Cu_3O_{7-x}$ sample (both in arbitrary units on a log scale).

higher temperatures could be due to the time difference in the microwave and dc resistance measurements. The similarities to those observed at the 90 K transitions, however, offer further evidence of a still higher-temperature (but unstable) superconducting transition such as reported for certain Y-Ba-Cu oxide samples from dc resistance measurements[5–7] and the reverse ac Josephson effect.[8]

Figure 4 shows the effect of a 5.5 T magnetic field on the 90 K transition in a $YBa_2Cu_3O_{7-x}$ sample. As can be seen, the magnetic field decreases the temperature at which the sudden increase in transmitted power occurs by about 3 K. It also decreases the sharpness of the power increase and continues to suppress the overall transmission coefficient even at temperatures far below T_c. We have also looked carefully for the effect of a magnetic field on the step occurring around $T = 265$ K. Such a field dependence could provide strong evidence that this step is also associated with a superconducting transition. Any such dependence, if it exists, is smaller than that observed at 90 K. Unfortunately, the present stability for our spectrometer and the reproducibility of data taken in this region are not sufficient to show unambiguously that a field of 5.5 T has a definite effect.

In Fig. 5 the temperature dependence of the transmitted power for a $La_{1.8}Ba_{0.2}CuO_{4-x}$ sample is plotted for both zero field and an applied field of 5.4 T. Also shown in the figure is the dc resistance which was measured in zero field. The features seen in Fig. 5 are very similar to those observed in the Y-Ba-Cu oxide samples. For temperatures below T_c ($\cong 30$ K), the transmitted power once again increases quite rapidly. The applied magnetic field

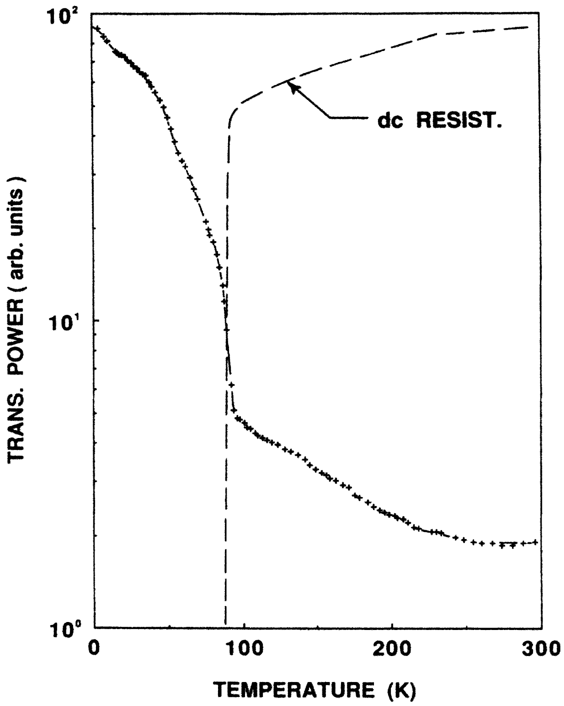

FIG. 3. Temperature dependence of the transmitted microwave power and dc electrical resistivity of a $Y_5Bu_6Cu_{11}O_x$ sample.

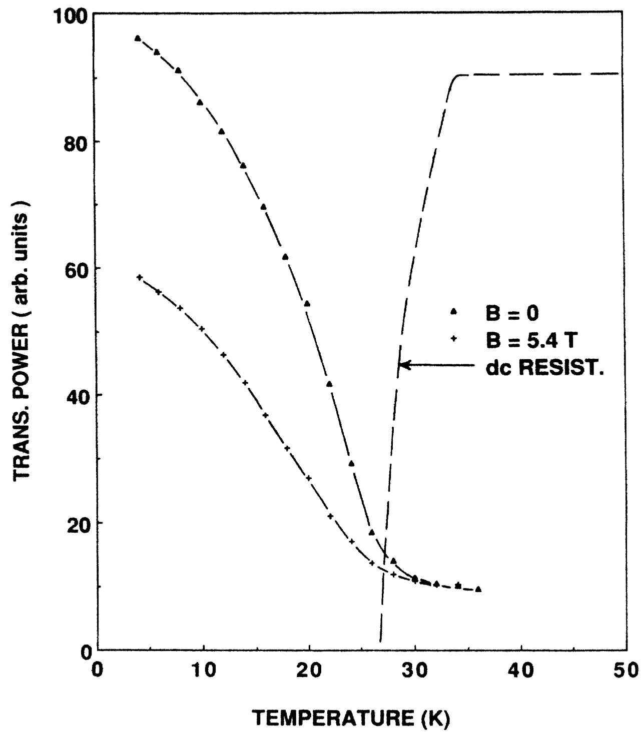

FIG. 5. Temperature dependence of the transmitted microwave power and dc electrical resistance of a $La_{1.8}Ba_{0.2}CuO_{4-x}$ sample. Also plotted is the transmitted power for an applied field of 5.4 T.

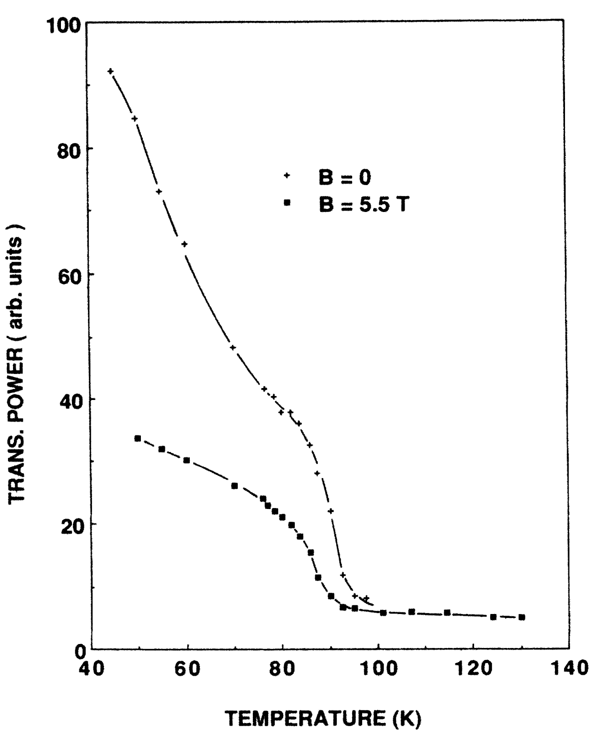

FIG. 4. Effect of a magnetic field on the temperature dependence of the transmitted microwave power for a $YBa_2Cu_3O_{7-x}$ sample.

suppresses the transmission of power at all temperatures below T_c and the sharpness of the power increment in the vicinity of T_c. By comparing Figs. 4 and 5, it is seen that a magnetic field has a similar effect on both the La-Ba-Cu-O and Y-Ba-Cu-O samples, namely an increasing suppression of the transmitted power at lower temperatures. Just the opposite would be expected for a type-I bulk superconductor due to the saturation of the superconducting properties at temperatures far below T_c. The stronger magnetic field dependence observed here at lower temperature is most likely related to a weaker superconductivity associated with Josephson coupling between superconducting grains.

The transmitted power which we measure is related to the surface impedance of the sample at microwave frequencies. For microwave propagation in the TE_{10} mode in an open waveguide of length L the transmission coefficient for the microwave fields is given by[9]

$$T = \exp\left\{ -R_s L \left[1 + \frac{2b}{a} \left(\frac{f_c}{f} \right)^2 \right] / b \left(\frac{\mu_0}{\epsilon_0} \right)^{1/2} \times \left[1 - \left(\frac{f_c}{f} \right)^2 \right]^{1/2} \right\}, \quad (1)$$

where f is the frequency and R_s is the surface resistivity of the metal from which the waveguide is fabricated. Presumably, the signals we detect are described by an expression similar to Eq. (1) in which R_s is replaced by some type of weighted average of the surface resistances of the superconducting sample and the silver waveguide. The increase in the transmission coefficient below T_c is consistent with the decrease in R_s of the superconductor which occurs at this temperature. Because an exact relation for the transmitted field and the temperature dependence of R_s for silver are unavailable, and because some residual interference between waves from the top and bottom gaps remains, we have not attempted to fit our observations to theory. Nevertheless, it is clear from the data that there are a number of features in the temperature dependence of the transmission which correlated with the characteristics of the superconducting transitions and their dependence on magnetic field.

A better method of recording this type of data (which we are now pursuing) is one with the geometry uniquely defined in which the microwaves propagate through a "single slit" down the center of the sample, both halves of which are electrically bonded to the waveguide. In this case the microwave transmission depends only on the characteristics of the superconducting material itself. Such measurements (coupled with similar measurements of the reflection coefficient) appear to offer the possibility of a sensitive determination of the electrical and magnetic properties of this class of materials consisting of granular superconductors and coupled Josephson junctions. The advantage over more traditional techniques, which make use of a microwave cavity,[10,11] appears to be a higher sensitivity and the ability to study variations in R_s over a larger dynamic range.

This work was supported in part by National Science Foundation Grants No. DMR-84-14488 (G.L.D.) and No. DMR-84-00711 (L.E.W.). We are pleased to acknowledge helpful discussions with J. J. Chang and A. Janossy and technical assistance from R. E. Brauer.

[1]J. G. Bednorz and K. A. Müller, Z. Phys. B **64**, 189 (1986).
[2]M. K. Wu, J. R. Ashburn, C. J. Torng, P. H. Hor, R. L. Meng, L. Gao, Z. H. Huang, Y. Q. Wang, and C. W. Chu, Phys. Rev. Lett. **58**, 908 (1987).
[3]R. J. Cava, B. Batlogg, R. B. van Dover, D. W. Murphy, S. Sunshine, T. Siegrist, J. P. Remeika, E. A. Rietman, S. Zahurak, and G. P. Espinosa, Phys. Rev. Lett. **58**, 1676 (1987).
[4]G. L. Dunifer and M. R. Pattison, Phys. Rev. B **14**, 945 (1976).
[5]C. W. Chu, New York Times, March 10, 1987.
[6]L. C. Boume, M. L. Cohen, W. N. Creager, M. F. Crommie, A. M. Stacy, and A. Zettl, Phys. Lett. A **120**, 494 (1987).
[7]C. Y. Huang, L. J. Dries, P. H. Hor, R. L. Meng, C. W. Chu, and R. B. Frankel, Nature **328**, 403 (1987).
[8]J. T. Chen, L. E. Wenger, C. J. McEwan, and E. M. Logothetis, Phys. Rev. Lett. **58**, 1972 (1987).
[9]S. Ramo, J. R. Whinnery, and T. Van Duzer, *Fields and Waves in Communication Electronics* (Wiley, New York, 1965).
[10]W. P. Beyermann, B. Alavi, and G. Gruner, Phys. Rev. B **35**, 8826 (1987).
[11]R. Durny, J. Hautala, S. Ducharme, B. Lee, O. G. Symko, P. C. Taylor, D. J. Zheng, and J. A. Xu, Phys. Rev. B **36**, 2361 (1987). Figure 3 of this paper appears to show a temperature dependence of the microwave absorption which is opposite to our observations. The ordinate of the figure is actually a somewhat different quantity—the *derivative* of the microwave absorption with respect to magnetic field (obtained by field modulation). The temperature dependence of the absorption as determined from the cavity Q, however, is in agreement with our observations (P.C. Taylor, private communication).