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## Absorption at radio frequencies in superconducting $Y_1Ba_2Cu_3O_y$

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# Absorption at radio frequencies in superconducting $Y_1Ba_2Cu_3O_y$

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The large magnetic-field-dependent ac absorption in superconducting  $Y_1Ba_2Cu_3O_y$  ceramics and powders decreases slowly with increasing frequency in the range 2–16 MHz. The magnetic-field-dependent ac absorption is observed below  $T_c$  in superconducting  $Y_1Ba_2Cu_3O_y$  ceramics, powders and twinned crystals and in  $(La,Sr)_2CuO_4$  and  $BaPb_{0.75}Bi_{0.25}CuO_3$  powders and appears to contain distinct contributions from trapped flux and/or intragrain tunnel junctions and from ordinary superconductor surface impedance. The contribution from trapped flux and/or intragrain tunnel junctions is important in dc magnetic fields of up to approximately 20 G at all temperatures below  $T_c$ . There is an identifiable modulated ac absorption which changes slowly over more than 12 kG at low temperatures and which we propose is primarily caused by the ac surface impedance with possible contributions from the critical state. A twinned crystal of  $Y_1Ba_2Cu_3O_y$  exhibits the same ac absorption except that it has an anisotropic dependence on the applied dc magnetic field.

## I. INTRODUCTION

The recent discoveries of superconductivity at record high temperatures in copper-octahedral oxygen perovskites<sup>1</sup> have opened a new and exciting chapter in the field of superconductivity. While superconducting transition temperatures<sup>2</sup> of 125 K (and perhaps higher<sup>3</sup>) promise economical application of superconductivity on an unprecedented scale, the scientific community is striving to understand the mechanism and behavior of the new superconductors through experimentation and theoretical modeling. Many of the basic superconductivity-related properties of the new materials have been measured in recent months: the resistive transition and magnetic susceptibility,<sup>1</sup> critical fields,<sup>4</sup> the energy gap,<sup>5</sup> and macroscopic quantum phenomena.<sup>6</sup> We observed<sup>7</sup> an unusually large field-modulated microwave absorption signal below  $T_c$  in  $Y_1Ba_2Cu_3O_y$  ceramics and powders and have since observed<sup>8</sup> similar modulated absorption in a twinned crystal of  $Y_1Ba_2Cu_3O_y$  and in  $(La,Sr)_2CuO_4$  and  $BaPb_{1-x}Bi_xO_3$  (BPB) powders. The modulated absorption can be separated qualitatively and quantitatively into distinct contributions from the ac surface impedance observed in all superconductors plus a new contribution observed only at low dc field ( $< 20$  G) in the perovskite superconductors. In Ref. 7 we showed that the field-modulated ac absorption signal in  $Y_1Ba_2Cu_3O_y$  at 9.4 GHz appears abruptly at  $T_c$  as a sample is cooled and increases steadily to a constant value at low temperatures. The modulated absorption signal at a fixed temperature decreases as a dc magnetic field is applied, decreasing rapidly over approximately 20 G (the contribution which appears unique to perovskite superconductors) and then more slowly over several kG. We now report the frequency dependence of the field-modulated ac absorption component of the macroscopic ac magnetic sus-

ceptibility in superconducting  $Y_1Ba_2Cu_3O_y$  powders measured at temperatures from 20 K to  $T_c$  and frequencies 2–16 MHz as well as at 9.4 GHz. We observe a similar modulated ac absorption at 9.4 GHz at low fields in superconducting ceramics, powders, and twinned crystals of  $Y_1Ba_2Cu_3O_y$  and in superconducting powders of  $(La,Sr)_2CuO_4$  and BPB.

In the next section we describe how the ceramic samples were made and prepared. The following section describes the radio and microwave frequency measurements. There follows a detailed description of the behavior of field-modulated ac absorption under a variety of conditions: varying temperature, dc magnetic field, sample quality, and measurement frequency. We conclude with a discussion of several possible mechanisms and further details which should be included in a complete model of the ac absorption in high-temperature superconductors.

## II. SAMPLE PREPARATION

The superconducting  $Y_1Ba_2Cu_3O_y$  ceramic samples were prepared by solid-state reaction of  $BaCO_3$  (reagent grade),  $Y_2O_3$  (99.999%), and  $CuO$  (puratronic grade). All powders were mixed and ground for 24 h with ethanol and alumina. Mixed powders were dried and fired in an alumina crucible at 940 °C in air for 50 h. Subsequently, calcined powder was milled for 24 h and binder added. Pellets with 1.5 cm diameter and 0.3 cm thickness were pressed from the powder-binder mixture at 35 psi. The pellets were fired at 970 °C for 24 h and cooled at a rate of 50 °C/h. (The binder was completely removed during sintering.) The pellets were then annealed in oxygen at 15 psi at 950 °C and then cooled at a rate of 30 °C/h.

For the resistance measurements, leads were attached to the ceramic pellets with silver paint in a four-terminal con-

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figuration. The  $R(T)$  curves were not measurably affected by currents up to  $0.2 \text{ A/cm}^2$ .

Powdered samples were obtained by coarsely grinding ceramic pieces (e.g., particles range from  $< 1 \mu\text{m}$  to several dozen  $\mu\text{m}$  in size). The twinned crystal was selected from the powder and glued to the end of a quartz rod. The samples studied by ac absorption, ceramic pieces, powders, or the twinned crystal, were placed in a 2 mm diameter  $\times$  250 mm long ESR grade quartz tube. The samples were flushed and filled with helium gas at atmospheric pressure, and sealed to prevent sample deterioration from contact with oxygen and water vapor.

### III. EXPERIMENTAL TECHNIQUE

The modulated ac absorption measurements were made with the two different types of apparatus described below. The modulated absorption signal is conceptually the same in both types of apparatus. The sample was subjected to three magnetic fields: a dc field which determines the state of the sample (at fixed temperature); a low-frequency (350 Hz or 100 kHz) field which modulates the sample state a small amount about the dc state; and a high-frequency (2 MHz–9.4 GHz) probing field which induces a magnetic response in the sample which was detected as a function of the lower-frequency modulation.

The microwave (9.4-GHz) modulated ac absorption measurements were made in the tuned resonant cavity (Fig. 1(a)) of a Brüker ER200D-SRC ESR spectrometer and the measurements made in the same manner as ordinary ESR spectra except that in this case the measured absorption is not from a magnetic resonance. The quartz tube containing the superconducting sample was placed in a position of max-

imum microwave magnetic field (minimum electric field) in the resonant cavity [see Fig. 1(a)] which was critically coupled (minimum reflectivity) to the input/output waveguide. A dc magnetic field plus a small ac modulation field (at 100 kHz) were applied and the modulated part of the reflected power from the resonant cavity was detected by phase-sensitive amplification at the frequency of the modulation field. The demodulated signal was then digitized and recorded by a computer. The field modulation technique used gives rise to a derivative-like signal proportional to the imaginary part (absorption) of the macroscopic ac magnetic susceptibility of the sample when the modulation amplitude exceeded approximately 1 G. Lower modulation amplitudes resulted in dramatically different results, depending on the direction of the dc field scan.

The radio frequency (rf) modulated absorption measurements were performed with the same helium flow cryostat but with a different ac susceptometer. Figure 1(b) shows the arrangement of the superconducting sample and twin rf coils of the Varian model V-4200B NMR spectrometer. The rf waves were generated by a tuned circuit containing the "transmitter" coil and detected by an identical "receiver" coil and circuit oriented nearly orthogonal to the "transmitter" coil. The received power was then proportional to the mutual inductance of the coils. The (macroscopic) ac magnetic susceptibility of the sample placed within the coils [Fig. 1(b)] altered their mutual inductance. The relative phase of the "transmitter" and receiver circuits could be adjusted so that either the in-phase (dispersion) or out-of-phase (absorption) components of the macroscopic ac magnetic susceptibility could be measured. A small ac modulation magnetic field (0.2–2.0 G at 355 Hz) plus a dc magnetic field (0–12 kG) was applied to the sample and the modulated (at 355 Hz) part of the receiver power detected by phase-sensitive amplification and recorded by a computer as in the microwave absorption measurements. The modulated ac absorption signal in the rf apparatus includes both magnetic and electric contributions, unlike the microwave modulated absorption signal which includes magnetic contributions only. Because of the similarity of the radio and microwave frequency signals, we assume they are both magnetic in origin.

In both the microwave and rf ac absorption measurements, a standard ESR resonant medium (DPPH) was used to calibrate the magnitude of the absorption measured in the superconductor. This is particularly important in order to compare the signal strength at different frequencies as the mutual inductance and detector efficiency of the rf coils are a strong function of frequency. Care was taken to minimize the effect of the sample on coil or cavity tuning.

Temperature control during the ac absorption measurements was accomplished with an Air Products Heli-Tran helium flow cryostat with calibrated chromel-gold (0.07% Fe) thermocouples and microprocessor feedback to a heater mounted in the helium flow [see Fig. 1(a)]. Since the thermocouple was outside the sample tube, a small systematic error in temperature was unavoidable; the superconducting sample was generally a few degrees warmer than the thermocouple mounted upstream in the helium gas flow.

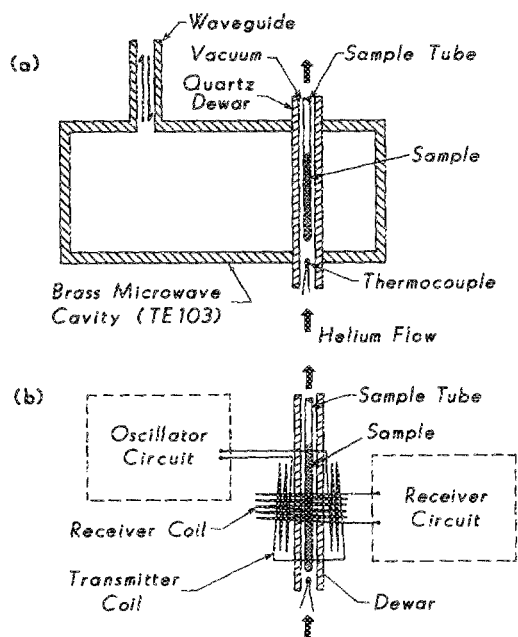


FIG. 1. Diagram of the ac susceptibility apparatus: (a) Microwave resonant cavity and helium flow cryostat used at 9.4 GHz. (b) Matched rf coils and helium flow cryostat used in the range 2–16 MHz.

Because of the number and variety of reports of similar but different measurements of ac absorption in perovskite superconductors the casual reader, and perhaps even the expert, may have trouble sorting out the similarities and the differences. The most common observation reported is that of a "zero-field" absorption signal obtained with a field modulation technique in an electron-spin-resonance apparatus like the Bruker ESR spectrometer used in our microwave frequency measurements. Published reports of this nature of which we are aware are Refs. 7-13 though many new reports appear in the literature each month. The use of an ESR spectrometer benefits from extremely high sensitivity but the large diamagnetic dispersion response of superconducting samples can produce false absorption signals as pointed out in Ref. 10.

A second, similar technique in studying the ac magnetic response of superconductors is measurement of the frequency shift (dispersion) and quality factor (absorption) of a resonant cavity,<sup>13-16</sup> or of a waveguide<sup>17</sup> containing a superconducting sample *without* an additional modulation magnetic field as used in ESR spectrometers.

The techniques involving ESR spectrometers have some important variations. Modulation amplitude appears to be the most important parameter; Portis *et al.*,<sup>13</sup> Blazey *et al.*,<sup>11</sup>

and Durny *et al.*<sup>18</sup> report a dramatically different modulated signal, which depends antisymmetrically on the *direction* of the dc field scan, observed at small modulation amplitudes. The radio frequency measurements reported here are at relatively large modulation amplitudes,  $\frac{1}{2}$  G, and show no hysteresis or dependence on the scan direction. For this and other reasons to be discussed, we observe that *modulation techniques generally do not allow the superconducting samples to be in equilibrium.*

#### IV. RESULTS

We will first characterize the behavior of the ac absorption and show that it includes a contribution at low dc fields unique to the high  $T_c$  perovskite superconductors, behaving differently from the commonly observed "skin effect" surface impedance (absorption) due to conduction electrons. First, the observed ac absorption has a pronounced dependence on the magnitude of an applied dc magnetic field. Figure 2 shows typical absorption signals observed in superconducting  $Y_1Ba_2Cu_3O_x$  powder at 6 MHz. There are two distinct signals antisymmetric about zero magnetic field. The first is a relatively narrow "derivativelike" signal [Fig. 2(a)] with a width of approximately 10-30 G, depending on

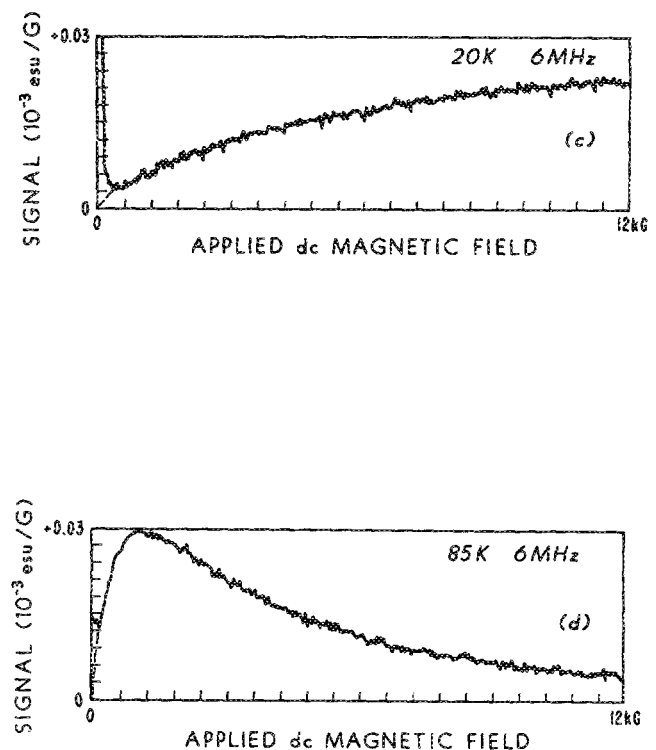
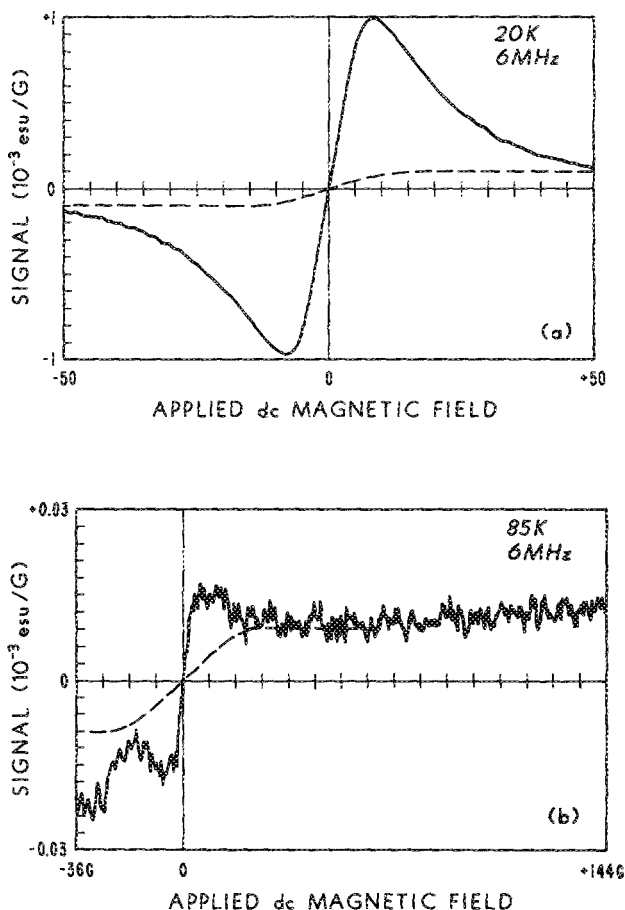


FIG. 2. Magnetic field dependence of the modulated ac absorption in  $Y_1Ba_2Cu_3O_x$ . (a) At 20 K between -100 and +100 G. (b) At 85 K from -36 to 144 G. (c) and (d) From 0 to 1.2 T at 20 and 85 K, respectively. The dotted lines show the approximate contribution from the surface impedance.

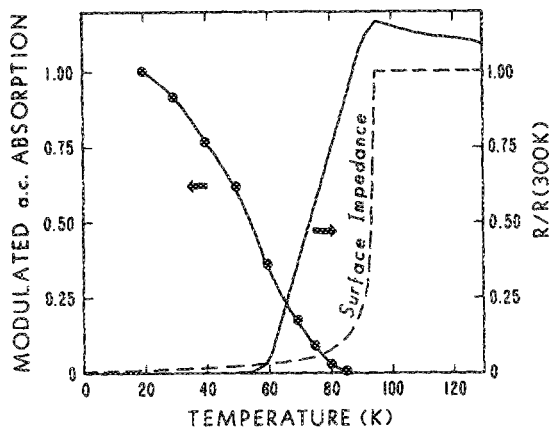


FIG. 3. Temperature dependence of the modulated ac absorption signal at 6 MHz (relative to the signal at 20 K) and the dc resistivity (relative to the resistivity at 300 K). The usual surface impedance is shown schematically by the dashed line.

temperature and frequency. The narrow "signal" sits on top of a very broad signal indicated by the dotted line in Fig. 2(a). As the temperature approaches  $T_c$  from below the narrow contribution to the modulated absorption decreases more rapidly than the broad contribution as is evident in comparing Figs. 2(a) and 2(b). The results and discussion which follow refer to the narrow, or low-field, modulated absorption signal unless otherwise noted. The ac dispersion measurements from 2 to 16 MHz result in a signal similar to those in Fig. 2 except that the dispersion has twice the width and half the magnitude (relative to the standard DPPH) of the modulated ac absorption.

The pronounced temperature dependence of the narrow modulated absorption is shown in Fig. 3. The ac absorption appears only at temperatures below  $T_c$ , the superconducting transition temperature, and increases steadily to a large, constant, value at low temperatures.<sup>7</sup> The modulated absorption appears *concurrently with presence of the superconducting state*. The high sensitivity of the modulated absorption measurements (especially at 9.4 GHz where the resonant cavity one of used has a dynamic range of at least five decades) greatly exceeds that for resistive or Meissner flux exclusion measurements near  $T_c$ . The modulated ac absorption is therefore one of the most sensitive measure of  $T_c$  presently available in ceramics and twinned crystals.<sup>7,12</sup> The temperature dependence of the ac surface impedance<sup>16</sup> ("skin-effect") in a superconducting  $Y_1Ba_2Cu_3O_7$  is also shown in Fig. 3 for comparison. The surface impedance in  $Y_1Ba_2Cu_3O_7$  is weaker than the surface impedance in most metals due to the low carrier density of about  $10^{21} \text{ cm}^{-3}$ . Another indication of the unique, superconductivity-related character of the ac absorption is the dependence on sample quality. The modulated ac absorption signal, as well as the observed Meissner fraction, increases with the increasing proportion of superconducting  $Y_1Ba_2Cu_3O_7$  in the sample. Even samples with 100%  $Y_1Ba_2Cu_3O_7$ , the strength of the ac absorption may still vary with slight variations in preparation conditions.

Powders of  $La_{1.9}Sr_{0.1}CuO_4$  and  $BaPb_{0.75}Bi_{0.25}O_3$  (BPB) showed similar modulated absorption signals at 9.4

GHz as shown in Figs. 4(a) and 4(b). (The signals were too weak to measure with the radio frequency apparatus.) The modulation amplitude for both spectra was 2.5 G which was sufficiently large to produce an antisymmetric signal in  $La_{1.9}Sr_{0.1}CuO_4$  but not in BPB. The asymmetry observed in Fig. 4(a) is similar to that observed in  $Y_1Ba_2Cu_3O_7$  sample under similar conditions but with modulation amplitude  $\sim 1/4 \text{ G}$ .<sup>18</sup> The temperature dependence of the magnitude of the modulated absorption signal in powdered  $La_{1.85}Sr_{0.15}CuO_4$  is shown in Fig. 4(c).

The frequency dependence of the modulated ac absorption in a powder of  $Y_1Ba_2Cu_3O_7$  is shown in Fig. 5. The

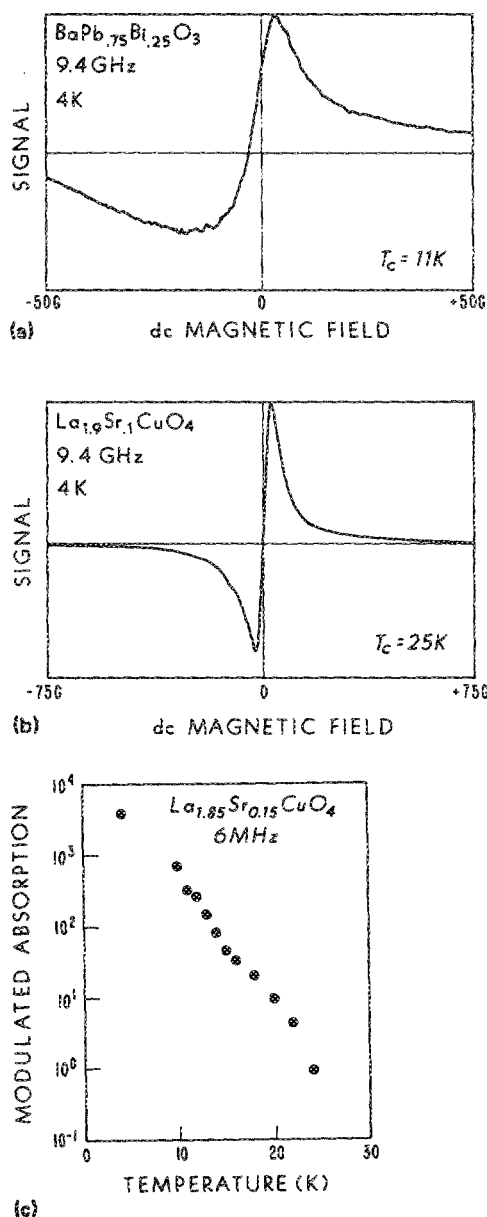


FIG. 4. Modulated ac absorption signals at 9.4 GHz at 4 K in (a)  $BaPb_{0.75}Bi_{0.25}O_3$  ( $T_c \approx 11 \text{ K}$ ) with modulation amplitude 2.5 G and (b)  $La_{1.9}Sr_{0.1}CuO_4$  ( $T_c \approx 25 \text{ K}$ ) with modulation amplitude 2.5 G. (c) Magnitude of the modulated absorption in  $La_{1.85}Sr_{0.15}CuO_4$  powder vs temperature.

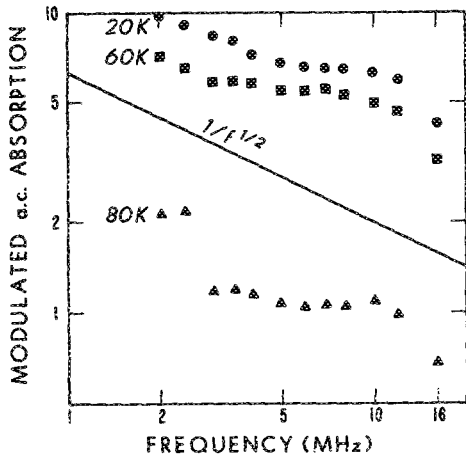


FIG. 5. Frequency dependence of the modulated ac absorption from 2 to 16 MHz at three different temperatures. The solid line is the function  $f^{-1/2}$  shown for comparison.

radio frequency (2–16 MHz) measurements were made using the nearly orthogonal tuned coils as described above [Fig. 1(b)]. The modulated absorption at a fixed temperature decreases as the frequency increases. The solid line has a slope of  $-\frac{1}{2}$  and is slightly steeper than the data. The modulated absorption (narrow peak) at 9.4 GHz at 20 K is approximately 40 times weaker than at 10 MHz for an overall change of 1.7 decades with a frequency change of nearly 4 decades. The surface impedance,<sup>15,16</sup> in contrast, increases approximately as  $f^2$ .

Figure 6(a) shows the temperature dependence of the modulated absorption strength and the modulated dispersion signals at several frequencies. The widths of the narrow modulated absorption and dispersion signals at different frequencies are shown in Fig. 6(b).

Though the ac absorption (see Fig. 3) is not significantly different in superconducting ceramic pieces or powders, when a crystal which is presumably twinned is selected from the powder the observed signal exhibits two distinctly different peaks depending on how the sample is oriented with respect to the applied dc magnetic field. A  $100 \times 100 \times 10 \mu\text{m}^3$  twinned crystal plate of  $\text{Y}_1\text{Ba}_2\text{Cu}_3\text{O}_7$  was mounted with the  $c$ -axis perpendicular to the resonant cavity axis [Fig. 1(a)]. The microwave absorption was measured for different orientations of the applied dc magnetic field. The narrow ac absorption signature appeared as two peaks with different widths (Fig. 7). When the applied field was oriented parallel to the crystal  $c$  axis (also perpendicular to the plate), a signal with 8 G width was observed while when the applied field was oriented in the  $a$ - $b$  plane (also the plane of the plate), the width was about 40 G. (The sample was cooled in a field of approximately 25 G, the residual field of iron-core magnet used, and this may have resulted in increased overall line widths.<sup>18</sup>) A powder or ceramic piece exhibits a signal which is the rotational average of these two signals, about 20 G wide (when cooled in a 25-G field). There are three possible sources of the uniaxial symmetry shown in Fig. 7. The first possibility is that the absorption is uniaxial in the crystal

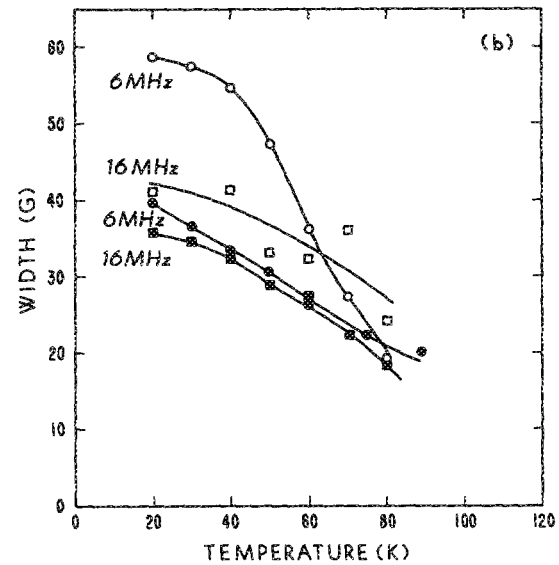
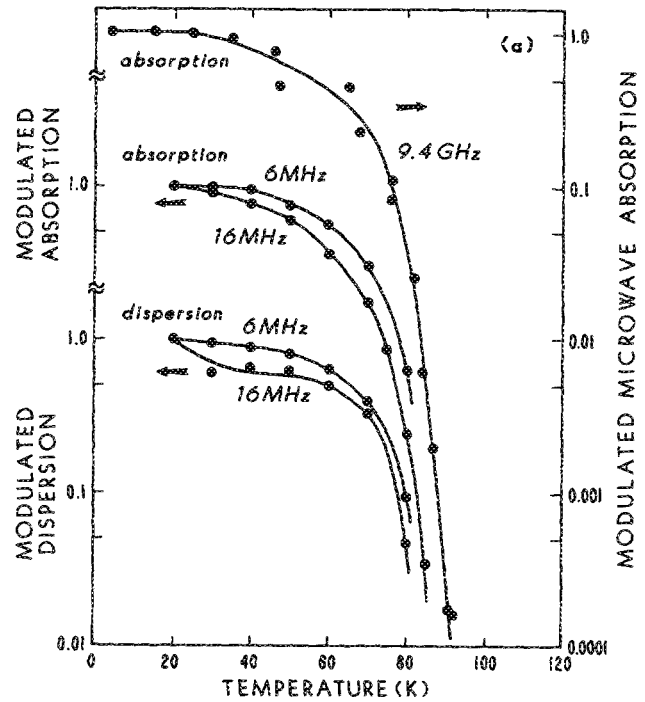


FIG. 6. Temperature dependence of the magnitude (a) and width (b) of the modulated ac dispersion and absorption at different frequencies. The data at each frequency are individually normalized to the signal at 20 K. The solid lines are guides to the eye. The open (closed) symbols in (b) represent the dispersion (absorption) signals.

lattice orientation (i.e., with respect to the  $c$  axis).<sup>19</sup> The crystal geometry could also account for the two peak widths; when the applied field is perpendicular to the plate, flux is more readily forced into the sample even at fields below  $H_{c1}$ . The third possibility is the anisotropy which would appear if twinning planes affect the ac absorption. But twinning planes are usually oriented perpendicular to crystal plates and would show a different anisotropy axis than that observed. We do not know if the sample studied has twinning planes, or how they are oriented, but reports of studies on many crystals of  $\text{Y}_1\text{Ba}_2\text{Cu}_3\text{O}_7$  deduce that the twin planes are preferentially oriented perpendicular to a plate.<sup>20</sup>

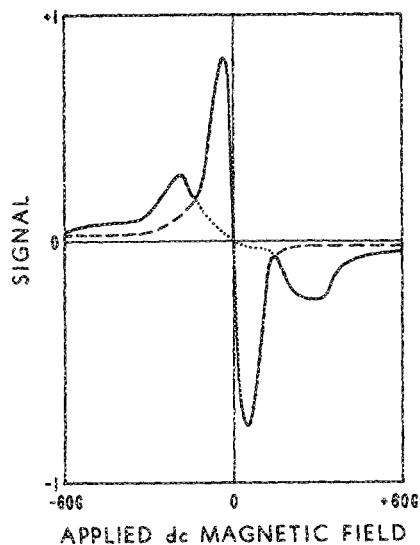


FIG. 7. Uniaxial ac absorption signal of a twinned crystal plate ( $100 \times 100 \times 10 \mu\text{m}^3$ ) at 4 K, 9.4 GHz. The narrowed ( $\sim 8$  G) signal appeared only when  $H \perp$  plate and the wide ( $\sim 40$  G) peak primarily when  $H \parallel$  plate. The modulation amplitude was 2 G.

Both the crystal and the powders exhibit significant hysteresis and field-cooling effects, as was observed by Müller *et al.*<sup>21</sup> in  $(\text{La,Sr})_2\text{CuO}_4$  in the low-frequency magnetic susceptibility. Our observations<sup>18</sup> show that cooling in fields as small as a few Gauss causes the narrow absorption signal [Fig. 2(a)] to broaden and shift its center to a nonzero applied field (e.g., flux is trapped in the powders or ceramic pieces during cooling). The shift in the signal center is then a direct measure of the amount of trapped flux and the broadening a measure of the nonuniform fields in the sample due to the trapped flux. A similar effect is obtained even in a sample cooled in zero field if dc magnetic field of a few kG or more is applied at low temperature and then removed.<sup>21-23</sup> The effects of cooling  $\text{Y}_1\text{Ba}_2\text{Cu}_3\text{O}_7$  in a field or applying a large field at low temperatures occur only below a "reversibility" temperature ( $T^*$  in Ref. 21) of about 80 K.<sup>18</sup>

Two other observations correlate well with the modulated ac absorption measurements. A dc voltage up to a few dozen microvolts appeared across ceramic samples of  $\text{Y}_1\text{Ba}_2\text{Cu}_3\text{O}_7$  when irradiated with 9.4-GHz microwaves but only at temperatures below  $T_c$ .<sup>24</sup> The dc voltage scaled linearly with microwave power up to a characteristic saturation power and decreased to one-half its zero field value at about 20 G, similar in shape to the integral of Fig. 2(a). We suspect the induced voltage may be the result of ac Josephson rectification by the many grain boundaries and the twins within each grain but added together randomly due to the random phases of the individual junctions. The modulated ac absorption signal width would then correspond to the distribution of critical fields of the junctions, not the bulk critical field of the material. The second observation related to Josephson junctions is in the behavior of the critical current of a thin bridge of  $\text{Y}_1\text{Ba}_2\text{Cu}_3\text{O}_7$  ceramic in dc magnetic fields and microwave radiation. The critical current decreases rapidly as a magnetic field is applied, again with a

width of about 20 G as in Fig. 2(a), but with faint oscillations like those observed in a single Josephson junction<sup>25</sup> smeared out due to the presence of many junctions.

## V. DISCUSSION

There are several characteristics of the unusually large modulated ac absorption which should be taken into consideration when evaluating potential mechanisms: (1) the sensitivity to small magnetic fields, (2) the precipitous increase in the magnitude of the modulated absorption below  $T_c$ , and (3) the frequency dependence. The other observations described in the previous section: uniaxial signals observed in a twinned crystal, hysteresis in a magnetic field, and evidence for ac Josephson effects are important clues to the mechanism of ac absorption. We will now discuss four distinct mechanisms for the modulated ac absorption and weigh their predictions against our observations. These mechanisms are dissipation in an ensemble of current loops with glassy behavior, fluxons in long Josephson junctions, pinned flux tubes in the critical state, and surface impedance.

Broad resistive transitions observed in the high  $T_c$  ceramics have frequently been attributed to various mechanisms, one of them being glassy behavior among weakly coupled grains.<sup>1,21,26</sup> It is natural to interpret other observed properties in terms of a glassy state: magnetic hysteresis,<sup>21</sup> the field dependence of critical currents,<sup>6,27</sup> and the modulated ac absorption.<sup>23</sup>

Nonequilibrium behavior of disordered superconductors has often been attributed to a glassy state. The observed modulated ac absorption could also be described by a glassy state if it includes a distribution of current loops or paths *within or between* the twinned grains and includes a dissipation mechanism such as intragrain Josephson junctions which break up the current loops (Fig. 8). Just such a model has been proposed by Deutscher and Müller<sup>27</sup> in which the junctions with a superconductor-insulator-superconductor (SIS) structure consisting of a tetragonal insulating layer which is a single unit cell in width separating the misoriented orthorhombic superconducting material. The key assumption made in order to make this structure a true SIS junction is that the superconducting energy gap in the orthorhombic

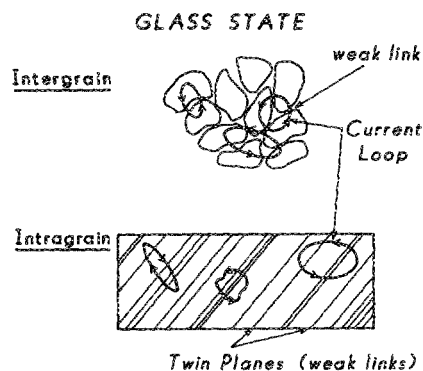


FIG. 8. Schematic diagram of current loops for a model of intergrain or intragrain glassy behavior.



material decreases dramatically at the S-I interfaces, thus producing a tunneling barrier. This process is possible, these authors propose,<sup>27</sup> because of the short superconducting coherence length ( $\sim$  lattice parameter) observed in the high  $T_c$  superconductors. At low temperatures the gap at the S-I interface would return to the bulk value  $\Delta(0)$  by the proximity effect<sup>28</sup> and hence the number of junctions would be reduced (see also Fig. 1 of Ref. 27). The modulated ac absorption, in contrast, does not decrease at low temperatures, but rises to an approximately constant value as shown in Figs. 3, 4, and 6. The requisite junctions, then, cannot be derived from the boundary-gap depression model,<sup>27</sup> but must arise in another manner. The model based upon a superconducting glassy state proposed in Ref. 27 could still apply, together with the existence of a "geometric critical field"  $H_{c1}^* = \phi_0/2s$ , where  $\phi_0$  is the flux quantum  $hc/2e$  and  $s$  is a typical loop area.

The temperature, frequency, and field dependencies are qualitatively consistent with this type of multiple-loop glassy behavior. As discussed in Ref. 27, a superconducting glassy state with typical current loop areas comparable to the grain size ( $\sim$  square micrometer) would have a critical field  $H_{c1}^* \approx 10$  G which is consistent with our measurements of the "width" of the modulated absorption feature (Fig. 6). The abrupt onset of the modulated absorption at  $T_c$  requires that the loss mechanism be associated with the superconducting state. Finally, the frequency dependence (Fig. 5) is qualitatively consistent with losses in a superconducting glassy state<sup>26</sup>; as the frequency increases, the larger current loops have too much self-inductance and cease to follow the driving field so that the overall absorption would decrease.

Let us assume that there are indeed Josephson junctions within each grain. Then a mechanism we propose<sup>8,18,24,29</sup> for the modulated ac absorption is the dissipative motion of Josephson fluxons in the junctions. Portis and Blazey have worked out just such a model.<sup>30</sup> This model is substantiated by our observations and measurements: microwave rectification by a bulk ceramic sample, magnetic field dependence of the critical current, and anisotropy of the modulated absorption in a twinned crystal. We observe<sup>24</sup> microwave-induced voltages of about a microvolt per milliwatt of microwave power irradiating a bulk superconducting ceramic sample. Also, the superconducting critical current of a small ( $1 \times 1 \times 1$  mm<sup>3</sup>) bridge has approximately the same magnetic field dependence as that of the modulated ac absorption [Fig. 2(a)]. Note that the modulated ac absorption, microwave rectification, and the critical currents all decrease dramatically in magnetic fields of approximately 20 G. The sensitivity to small magnetic fields [below  $H_{c1}$  (Ref. 19)] is consistent with the presence of Josephson junctions within the twinned crystals. Many such junctions can have one of their dimensions larger than the Josephson penetration depth, thus allowing the creation of quantized vortices by the external magnetic field. Induced ac currents through the junction can lead to Lorentz forces on the vortices and dissipation as well as many other interesting phenomena.

The third mechanism for the modulated ac absorption is dissipation by pinned flux tubes independently, or collectively in the critical state. When cooling samples in small mag-

netic fields, we observe that the modulated absorption signal is measurably altered when the magnitude of the field applied during cooling exceeds a few Gauss. When samples are cooled in fields larger than a few Gauss, the modulated absorption signal is observed to weaken and broaden significantly<sup>18</sup> so that the magnetic fields of flux tubes affects the source of the very low-field modulated absorption if only by providing a highly inhomogeneous magnetic field.<sup>26</sup> But the flux tubes themselves, whether individually or collectively, may be absorbing the incident radiation as the individual pinned vortices sit in a damped potential well and would absorb at all frequencies. The model proposed by Portis *et al.*<sup>13</sup> accounts for collective absorption by fluxons in the critical state at fields above a few Gauss.

Yeshurun and Malozemoff<sup>31</sup> have proposed that the dc susceptibility and hysteresis in the high  $T_c$  superconductors may be attributed to the critical state, the nonequilibrium flux state of a "hard" type-II superconductor.<sup>32</sup> The principle features of critical state electrodynamics<sup>32</sup> are only partially consistent with our results though there is sufficient room for reconciling the apparent discrepancies: (1) The ac absorption in the critical state is constant above a low threshold frequency but below the gap, contrary to the slow decrease of the modulated absorption observed (Fig. 5) over a limited range of frequencies. (2) The ac absorption in the critical state should begin above the lower bulk critical field  $H_{c1}$ , about 50 G (500 G) along the  $c(a,b)$  axis<sup>19</sup> in  $Y_1Ba_2Cu_3O_7$ , and may indeed contribute to the absorption at higher fields (see Fig. 9). Therefore, we propose that the

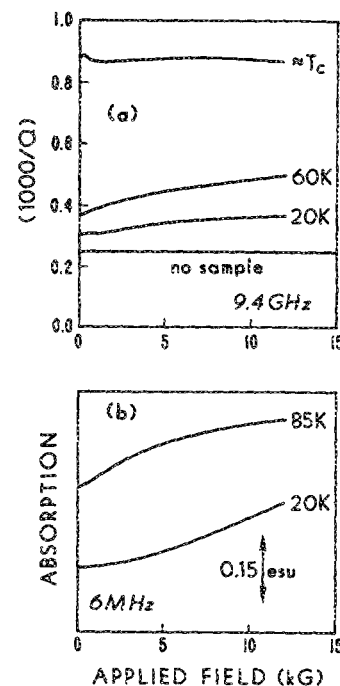


FIG. 9. (a) The inverse quality factor ( $1/Q$ ) of the microwave cavity containing a sample of  $Y_1Ba_2Cu_3O_7$  (see Fig. 1) at three temperatures. The quality factor ( $Q$ ) was determined by measuring the half width of the cavity resonance and includes contributions for the cavity walls as well as from the sample. (b) The broad component of the modulated absorption at 5 MHz in  $Y_1Ba_2Cu_3O_7$ .

observed modulated ac absorption below approximately 20 G appears to exclude contributions from the critical state due to Abrikosov fluxons.

The evidence indicates that the majority of the modulated ac absorption originates from the individual superconducting grains. Blazey *et al.*<sup>33</sup> report a revealing set of measurements of the ac absorption in a single crystal of  $Y_1Ba_2Cu_3O_7$ , which has a great deal of structure below 10 G and is highly anisotropic. The shape and magnitude of the low-field ac absorption [Fig. 2(a)] is the same in a ceramic piece and a powder ground from the ceramic. The anisotropic ac absorption in a single grain taken from a ceramic piece (Fig. 7) is an additional confirmation that the absorption takes place within each grain or at its surface.

Ordinary surface impedance or "skin effect" ac absorption has already been discussed. The modulated absorption signal at large fields (100 G to 1.2 T) is consistent with the expected behavior due to the surface impedance.<sup>16</sup> To confirm this, we measured the width of the cavity dip ( $Q$ ) of the microwave cavity with the superconducting sample in place. Figure 9 shows the cavity dissipation ( $1/Q$ ) at three different temperatures for fields from 0 to 1.2 T. At  $T_c$ , the sample is normal and the  $Q$  shows no dependence (within experimental error) on the applied magnetic field. At lower temperatures the ac absorption due to the surface impedance is smallest at zero field and increases as the magnetic field penetrates the sample. Since the magnetic field penetration depth exceeds 1000 Å in the materials while the superconductor coherence length is less than 50 Å, the superconducting state should not be sensitive to the grain surfaces.

The observed modulated ac absorption is also distinct from the generally observed peak in the imaginary part of the magnetic susceptibility which is generally observed just below  $T_c$ . This peak in the magnetic ac absorption is due to inductive coupling of the induced supercurrents to quasiparticle currents.<sup>34</sup>

## VI. SUMMARY

The macroscopic ac magnetic susceptibility in the high transition temperature superconductors exhibits a large field-modulated absorption even at frequencies well below the superconducting energy gap and at temperatures below the superconducting transition temperature. The observed modulated absorption is sensitive to dc magnetic fields of a few Gauss and decreases with increasing frequency from 2 MHz to 9.4 GHz. The modulated absorption is characteristic of individual grains of superconducting material. Microwave rectification in ceramic samples of  $Y_1Ba_2Cu_3O_7$ , has similar characteristics to the modulated absorption and may be due to ac Josephson rectification. Several mechanisms are proposed but further theoretical modeling and experimentation is necessary before a complete model is possible.

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