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Magnetic-field-dependent microwave properties of $YBa_2Cu_3O_x$ single crystals

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Magnetic-field-dependent microwave properties of $YBa_2Cu_3O_x$ single crystals are investigated as a function of temperature, oxygen annealing, and modulation amplitude. Three distinct signals are observed as the temperature is decreased from T_c to 2.4 K. It is established that the relative intensities of the signals depend on the degree of oxygen annealing. For the best annealed samples, only the high-temperature (just below T_c) signal remains prominent. The low-temperature signals are shown to be related to an inhomogeneous oxygen content in incompletely annealed samples. The hysteresis width increases at lower temperatures, indicating larger flux trapping. The sample-rotation experiments demonstrate that the microwave absorption depends on the state of the magnetization of the sample. In experiments in which the modulation amplitude is varied, we find that the previously observed anomaly in the shape of the signal is due to a fourth type of signal which has a looplike form and is very sensitive to field reversals.

I. INTRODUCTION

The discovery of high-temperature oxide superconductors[1] has given rise to a number of phenomenologically new observations. Among them are some complex and intriguing magnetic-field-dependent microwave properties.[2–9] It was shown that the absorption of microwaves on the sample surface becomes field dependent below T_c. The signal (i.e., the variation of absorption with magnetic-field sweep) grows from a baseline, which is all that is observed above T_c, to a maximum a little below T_c. The sharpness of the signal rise is in agreement with the transition width observed in resistance measurements. Even for small samples, the microwave signal is very large and can easily be detected by standard microwave devices. Since no leads have to be put on the sample for microwave measurements, this technique has become a convenient method for detecting superconductivity in new samples.[10] Among other features of the microwave signals observed in high-T_c superconductors, one may point out hysteresis and relaxation effects. The former can permit one to study flux trapping, while the latter indicates a property that could be associated with the superconductive glassy state and a flux creep process.

Most of the microwave measurements made so far were on ceramic samples. These consist of small superconducting grains coupled by weak Josephson links which can greatly influence the observed microwave characteristics of the sample. In order to determine intrinsic microwave properties of a given high-T_c compound, it is necessary to perform measurements on single crystals. Also, any anisotropy of the signals is averaged out in polycrystalline ceramic samples, and a single crystal study is needed to reveal it. In a recent report,[9] we have shown that single crystals of $YBa_2Cu_3O_x$ yield microwave signals which have certain features distinct from those in ceramic samples. As the temperature was decreased from T_c to 2.4 K, three different signals were observed. In a very good superconducting sample, oxygen-annealed for 10 days, the three signals appeared in separated temperature intervals, and could be studied individually. The signals differed in widths and hysteresis properties, and had pronounced anisotropy when the crystal was rotated with respect to the dc magnetic field. The temperature dependence of the signals was remarkable. Starting from T_c, the widest signal, having no hysteresis, would appear. Its amplitude would peak at a few degrees below T_c, and then decrease rapidly. Similar behavior could be observed for the other two signals, but at lower temperatures. This unusual behavior was discussed in terms of the conventional flux flow model in type-II superconductors,[11,12] and in terms of a recently proposed model of Josephson junctions at the nonsuperconducting narrow regions within an imperfect crystal.[13] In this paper we present a more extensive account of our microwave study of $YBa_2Cu_3O_x$ single crystals. In particular, we present the results of measurements on a number of samples, and with varying degrees of oxygen annealing. They show that the two signals which appear at lower temperatures are related to inhomogeneities in the oxygen content and other crystal imperfections. On the contrary, the strong signal, which appears just below T_c, seems to persist even in the best superconducting samples. We also show some effects of flux trapping and provide evidence for a fourth type of signal which causes an anomalous dependence of the signal shape on the modulation amplitude.

In Sec. II we describe the experimental conditions of our measurements. The effect of oxygen annealing on the microwave properties of $YBa_2Cu_3O_x$ single crystals is described in Sec. III. Some interesting observations on flux trapping and its consequences for the microwave absorption are presented in Sec. IV. The dependence of the signals on the modulation amplitude is analyzed in Sec. V, and the occurrence of a new type of signal at lower temperatures is proposed. A discussion and conclusions are presented in Sec. VI.

II. EXPERIMENT

All of the measurements reported here were made using a Bruker ER-200D electron-spin-resonance (ESR) spectrometer. It should be stressed that the observed signals are not those of electron-spin transitions, but an ESR spectrometer is simply used as a sensitive microwave device. Since the signals observed in superconductors are centered at zero magnetic field, it is convenient to arrange for the capability of through-zero field sweep, which is not available on most commercial ESR spectrometers. For this purpose we have used an additional pair of coils mounted on the sides of the microwave cavity. A dc current in these coils provided up to 100 Oe of field opposite to that of the main magnet. Thus a field sweep from -100 to $+10$ kOe could be achieved.

We have used an X-band (9.3 GHz) microwave bridge and a TE$_{102}$ microwave cavity. The microwave field develops a standing wave with a maximum of its magnetic component in the center of the cavity where the sample is placed for measurements. The polarization of the microwave magnetic field H_1 is along a vertical axis (call it x, cf. Fig. 1) and is parallel to the flat surface of the sample. This axis was also used for sample rotations. We have typically worked at 14 dB attenuation below 200 mW microwave power. The dc magnetic field H_0 is along a horizontal axis (call it z), i.e., $\mathbf{H}_0 \perp \mathbf{H}_1$. We have used an amplitude modulation of the field H_0. One can consider it as a parallel (i.e., along the z axis) superimposed field $H_m \cos(\omega_m t)$, where H_m is the amplitude (usually about 1 G except unless stated differently below) and ω_m is the frequency of the modulation (usually $2\pi\times 100$ kHz). Since the microwave absorption depends on the applied magnetic field, the field modulation brings about the modulation of the microwave absorption, and the signal can be obtained upon phase sensitive detection at the fundamental of the modulation frequency. This procedure, which is commonly used in ESR, yields first-derivative signals of the actual microwave absorption curves in the absence of hysteresis and slow relaxation effects.

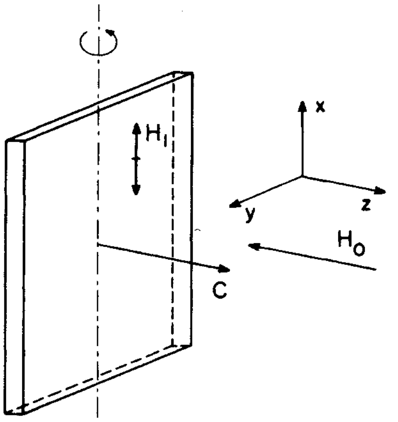

FIG. 1. Geometric arrangement of the magnetic fields and a crystal: H_0 is along the z axis, H_1 is parallel to the x axis. The sample is aligned with its c axis in the y-z plane (except as otherwise noted) and sample rotations are typically performed about the x axis.

The samples were prepared using the procedure described by Schneemeyer *et al.*[14] The typical dimensions of the sample were $2\times1\times0.03$ mm^3. The flat surface of the sample is parallel to the crystallographic *ab* plane. The samples were mounted on a holder and placed in an Oxford Instruments ESR-10 helium-gas-flow cryostat inside the microwave cavity. The temperature could be varied from 300 to 2.4 K.

III. SAMPLE AND OXYGEN VARIATIONS

In a previous report,[9] we have presented the temperature dependences of the microwave signals obtained in a well oxygen-annealed sample (10 days in oxygen at 490 °C) here referred to as sample I. The three signals, denoted as A, B, and C (see Ref. 9 and later sections of this paper for the form of these signals), appeared and disappeared consecutively as the temperature was decreased from T_c to 2.4 K. Here we present a more extensive study of the temperature dependences obtained in other samples, as well as in sample I after an additional annealing treatment.

Figure 2 shows the peak-to-peak signal amplitude for another sample also annealed for 10 days. The basic features are the same as for sample I reported previously.[9] However, it is found that the microwave technique can sensitively detect some differences, even between the samples prepared and treated in a similar manner. The signal denoted as A is now slightly shifted to lower temperatures, which implies a lower T_c and fewer good superconducting properties than in sample I. Note also that for sample II the signal of type B is as strong as the signal of type A, whereas in sample I it was an order of magnitude weaker than A. We have generally observed that signal B was much weaker than A in good superconducting samples,

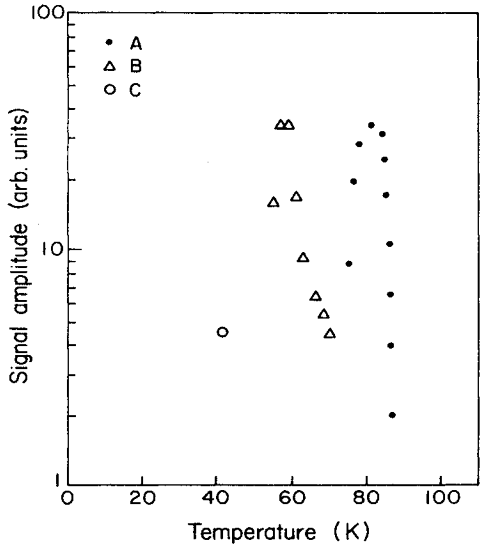

FIG. 2. Temperature dependence of the peak-to-peak amplitude of the three types of microwave signals (A, B, and C, as described in the text) in a single crystal of YBa$_2$Cu$_3$O$_x$ oxygen annealed for 10 days (sample II), recorded at $\mathbf{H}_0 \parallel \mathbf{c}$.

and it increased as the superconducting properties were degraded.

Figure 3 shows the temperature dependence of the signals observed in a sample oxygen annealed for 2 days, here referred to as sample III. The temperature dependence of signal *A* is particularly interesting. The first peak appears at 87 K. The signal then decreases and starts to rise again over a shoulder to a much larger peak (note the logarithmic scale) at 74 K. This result suggests that only some minor parts of sample III had a higher T_c, the same as most of sample I, but a major part of the sample had T_c suppressed by 10–15 K. This is most likely due to an inhomogeneity in the oxygen content throughout this well annealed sample. Single crystals of macroscopic dimensions along the *ab* plane are difficult to anneal properly because the diffusion of oxygen atoms must take place through long distances. Therefore, an inhomogeneous oxygen content throughout the sample is not a surprising result for insufficiently long annealing times and the microwave technique can nicely monitor it. One may notice also that signals *B* and *C* are relatively big in sample III.

Figure 4 shows the temperature variations of the signals in another sample, also oxygen annealed for 2 days (sample IV). One can see that T_c is also suppressed. However, there are no discernible small peaks and shoulders in the rise of signal *A*, but a gradual smooth increase. It seems that the oxygen content in sample IV has attained a different distribution than that in sample III. An important observation in sample IV is that signals *B* and *C* are not separated on the temperature scale (cf., Fig. 4). At some temperatures both signals can be observed superimposed as will be shown in the following section. This finding is important because it indicates that signals *B* do not evolve into signals *C* at lower temperatures, but the two signals seem to have independent origins.

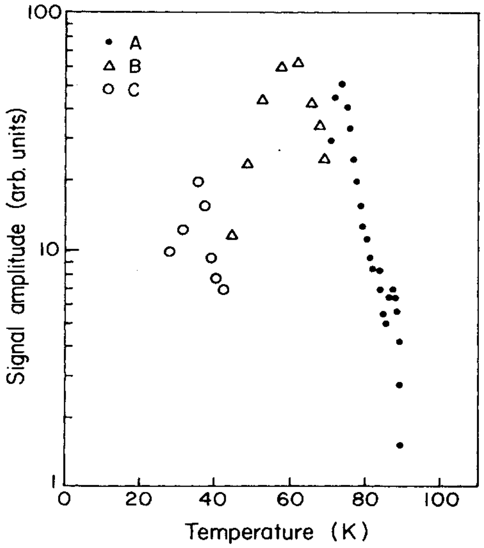

FIG. 3. Temperature dependence of the peak-to-peak amplitude of the three types of microwave signals (*A*, *B*, and *C*, as described in the text) in a single crystal of $YBa_2Cu_3O_x$ oxygen annealed for 2 days (sample III), recorded at $H_0 \| c$.

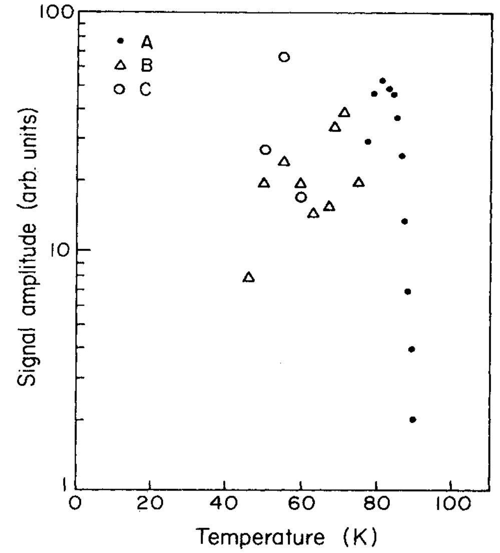

FIG. 4. Temperature dependence of the peak-to-peak amplitudes of the three types of microwave signals (*A*, *B*, and *C*, as described in the text) in a single crystal of $YBa_2Cu_3O_x$ oxygen annealed for 2 days (sample IV), recorded at $H_0 \| c$.

Finally, we have tested some "as-grown crystals" (i.e., with no additional oxygen annealing). They have been annealed only to the extent that some oxygen penetrates into the crystals during the cooling process in the furnace. An example of the temperature dependence of the signals in such a sample (here referred to as sample V) is shown in Fig. 5. One can see that T_c is suppressed by 15–25 K. Also, signals *A* and *B* overlap in their respective tempera-

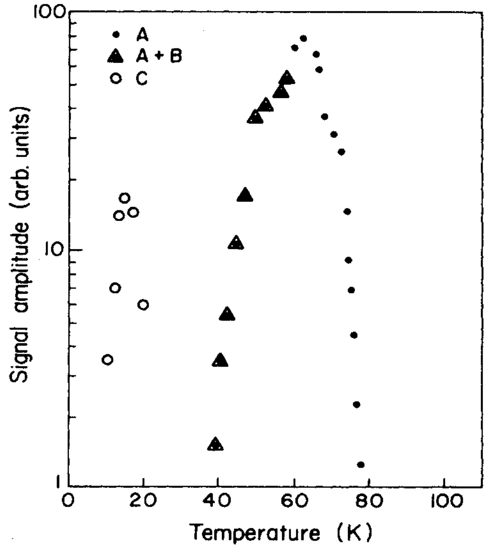

FIG. 5. Temperature dependence of the peak-to-peak amplitudes of the three types of microwave signals (*A*, *B*, and *C*, as described in the text) in an as-grown single crystal of $YBa_2Cu_3O_x$, oxygen annealed only during cooling to room temperature (sample V), recorded at $H_0 \| c$.

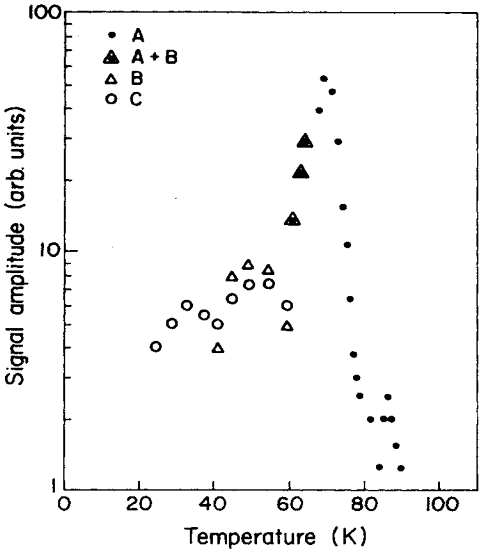

FIG. 6. Temperature dependence of the peak-to-peak amplitudes of the three types of microwave signals (*A*, *B*, and *C*, as described in the text) in a single crystal of $YBa_2Cu_3O_x$, denoted as sample I in the text, after vacuum annealing for 2 h. The recording was made at $H_0 \parallel c$.

ture ranges. At temperatures below the peak of signal *A*, one observes a gradual development of a hysteresis which is characteristic of signal *B*, but also a long tail characteristic of signal *A*. The two contributions are hard to separate with a reasonable certainty, and we present, therefore, the amplitude of the composite signal.

The above data show that there is a systematic trend in the change of the microwave signals with the oxygen annealing of the samples, but also that there are some differences between the samples treated in the same way. These latter are probably due to various defects associated with the single-crystal growth that can influence the subsequent oxygen-annealing process. In order to check that the above described differences between the samples are not exclusively due to defects, but depend crucially on the annealing, we have tried to vary the oxygen content in the same samples. Figure 6 shows the results obtained when sample I was vacuum annealed for 2 h at 490 °C. One can observe a small peak for signal *A* at 86 K, i.e., at the temperature where signal *A* previously had a peak.[9] A larger fraction of signal *A* is shifted to lower temperatures. It seems that vacuum annealing reduces sporadically the oxygen content so that an inhomogeneous distribution results. Signals *B* and *C* are increased in this process. These data are quite similar to those obtained for sample III. Further, vacuum annealing resulted in larger suppression of T_c, similar to that observed in sample IV, and beyond. Renewed oxygen annealing reverses the trend.

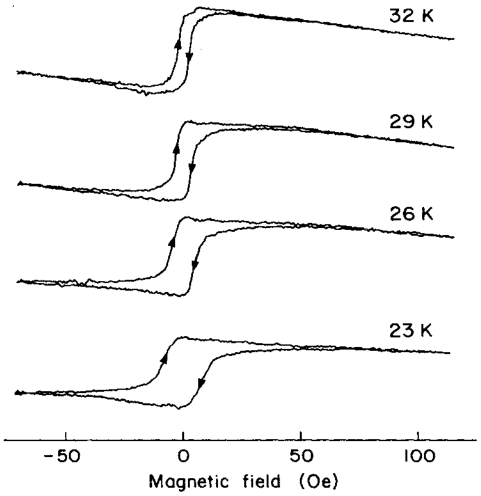

FIG. 7. Microwave signals of type *B* recorded in sample V after vacuum annealing for 2 h. The arrows indicate the sense of the hysteresis obtained in forward and reverse field sweeps. ($H_0 \parallel c$).

IV. HYSTERESIS AND FLUX TRAPPING

The occurrence of a hysteresis was one of the features used to make a distinction between the three microwave signals in $YBa_2Cu_3O_x$ single crystals.[9] In this section we report on some experimentally observed features of the hysteretic behavior of signals *B* and *C*. It was observed generally that the hysteresis width increased as the temperature was lowered. In some samples the signals overlapped in such a way that the hysteresis width of a given signal could not be determined precisely. However, in many cases it could be nicely followed. Such an example is shown in Fig. 7 for signal *B* obtained in sample V after vacuum annealing for 2 h. Signal *C* was not clearly seen in this particular case. In Fig. 8 we show a similar behav-

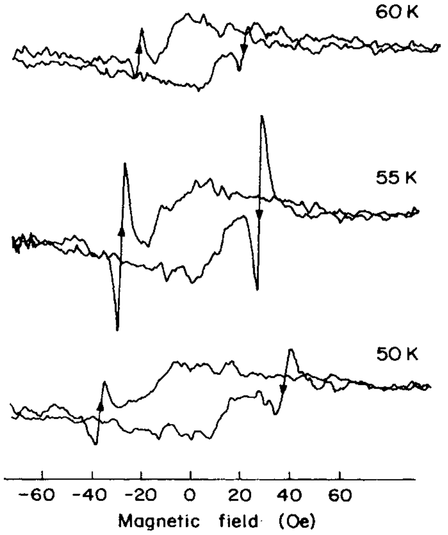

FIG. 8. Microwave signals of type *B* and *C* detected in sample IV (cf. also Fig. 4) at three different temperatures ($H_0 \parallel c$). The arrows indicate the traces recorded in forward and reverse field sweeps.

ior in sample IV where, as mentioned above, signals *B* and *C* were simultaneously observed. One can see how the hysteresis width increases for both signal *B* and signal *C*.

There is another characteristic feature of the hysteresis in the microwave signals of $YBa_2Cu_3O_x$ single crystals. At a given temperature the hysteresis width has some maximum value, detected when the magnetic-field sweep is made large enough. In other words, the hysteresis width cannot be increased by employing even larger field sweeps. However, one can record a reduced hysteresis width for smaller field sweeps. An illustration is shown in Fig. 9 where successive recordings of the signal are obtained with ever smaller field sweep ranges. The case presented is for sample IV, and the initial recording is the same as the middle one in Fig. 8. It is clearly seen that the hysteresis of both signal *B* and signal *C*, is reduced. The sharp line of signal *C* is always traced shortly after the field sweep is reversed on each side of the sweep range. Also note that the width of each line of signal *C* is considerably reduced. This indicates that signal *C* is inhomogeneous in its hysteresis, i.e., it consists of components with a slight distribution of hysteresis widths. At smaller field sweeps the hysteresis widths are reduced, and also their spread is minimized. In the limit of very small field sweeps, this procedure yields a single narrow line, centered at zero magnetic field, which is traced in both forward and reverse field sweeps. We have also noticed that in some cases this line shows an internal structure which is not related to the hysteresis effects.[9]

The appearance of a hysteresis is a sign of trapped flux in the sample. The present experiments indicate that the microwave absorption depends on the state of magnetization of the sample. An additional convincing proof is provided by the following experiment. In Fig. 10 we show a signal with a hysteresis. If the field is swept from the left, the upper curve is traced. The field sweep can be stopped when the recorder pen reaches point *P*, i.e., when the field is zero. If the sample is now rotated by 180°, the recorder pen shifts downward (i.e., the signal level changes) to point *P*′ on the other trace. Since the rotation was made in zero dc magnetic field, the only change that has occurred is in the sign of the magnetization of the sample with respect to the modulation field. The modulation field now modulates the magnetization that would have been created without the rotation if the field were swept from the right and stopped at point *P*′. Note that the same change in the sign of the signal would have been obtained if the phase of the modulation were changed by 180°. If the sample is again rotated by 180°, the signal level rises back to point *P*. Further rotations of the sample by 180° repeat the described cycle. A different behavior is experienced if the sweep starts from the left and stops at point *R* where the magnetic field is not zero. A rotation of the sample by 180° brings the signal level down to point *R*′, but subsequent rotations now leave the signal level unchanged. Also, if the sweep starts from the right and stops at *R*′ the rotations by 180° do not affect the signal level. This behavior can be explained in the following

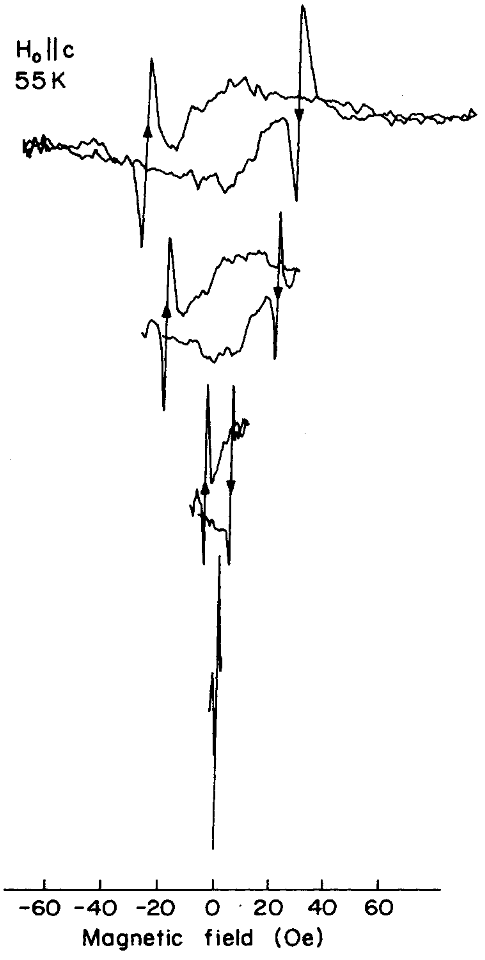

FIG. 9. Successive recordings of the signals in sample IV at 55 K (cf. Fig. 8) with reduced magnetic-field sweep ranges. The arrows indicate the sense of the traces.

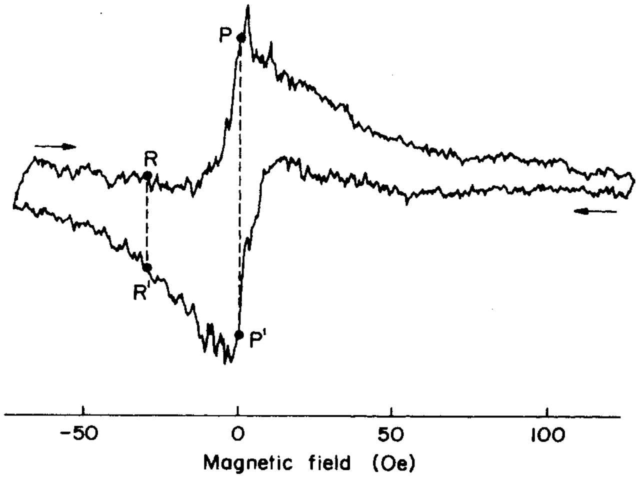

FIG. 10. Microwave signal with a hysteresis (sample I vacuum annealed for 2 h, 57 K, $\mathbf{H}_0 \| \mathbf{c}$). The arrows indicate the sense of the field sweep. Successive rotations of the sample by 180° in zero field change the signal level from *P* to *P*′ and back. In nonzero field the signal level changes from *R* to *R*′, but not back (see the text).

way. The sweep from the left to point R creates some magnetization in the sample. Since the rotation of the sample by 180° now occurs in a nonzero magnetic field, the magnetization is not simply rotated with the sample, but undergoes a change. The final magnetization is that which would have been created if the sample were at rest and the magnetic field were swept from the right (e.g., from a field opposite to that in point R) towards R'. In subsequent rotations the same process occurs, so that the signal level remains at R'. These experiments are very characteristic of these superconducting samples and show that the modulation field indeed affects the magnetization, which in turn, determines the microwave absorption.

V. EFFECTS OF MODULATION AMPLITUDE

It has been noticed in the microwave studies of ceramic samples that the shape of the signal could be varied with the modulation amplitude.[5,6] From this puzzling observation it was inferred that the signals detected by the use of magnetic-field modulation were not the derivatives of the actual absorption curves. We have carefully investigated this interesting phenomenon on the signals in single crystals. Figure 11 shows a series of signals of type A recorded with different modulation amplitudes. The receiver gain was varied inversely in order to preserve the magnitude of the detected signal. It can be seen that, to within the experimental error in noise, there is no effect of the modulation amplitude on the shape of signal A. Also, the amplitude of signal A depends linearly on the modulation amplitude. Only when the modulation amplitude exceeds the width of the rising step in the signal, does one start to observe a distortion of its shape. These are the characteristics of derivative signals.

However, at lower temperatures, where the hysteretic signals appear, we have found a similar dependence on the modulation amplitude as reported previously in ceramic samples.[5,6] Figures 12–14 show three series of signals taken with varying modulation amplitudes at different temperatures. The systematic evolution is the following. As the modulation level is decreased, the forward and reverse traces split to form elongated loops which taper towards higher fields. At lower temperatures the loops become larger for the same modulation amplitude. In the previous reports on the modulation amplitude effects,[5,6] it was assumed that the signal consisted of a single component. However, a closer inspection of the traces in Figs. 12–14 reveals that the signal may consist of two components. The first one yields steplike traces, while the second one tends to form elongated loops, which, for a limited field sweep range, take a rectangular form. From Figs. 12–14 one can see that the size of the step changes

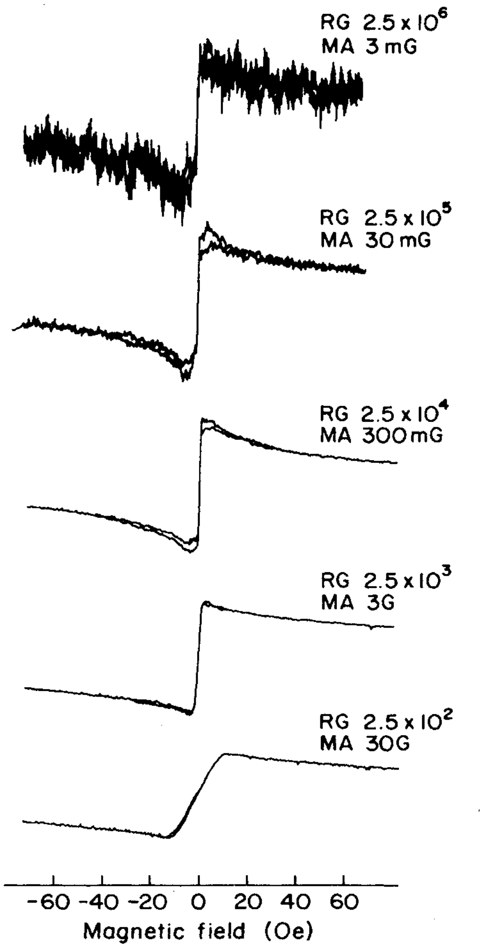

FIG. 11. Series of signals of type A recorded with variable modulation amplitude (MA) and compensating change in receiver gain (RG).

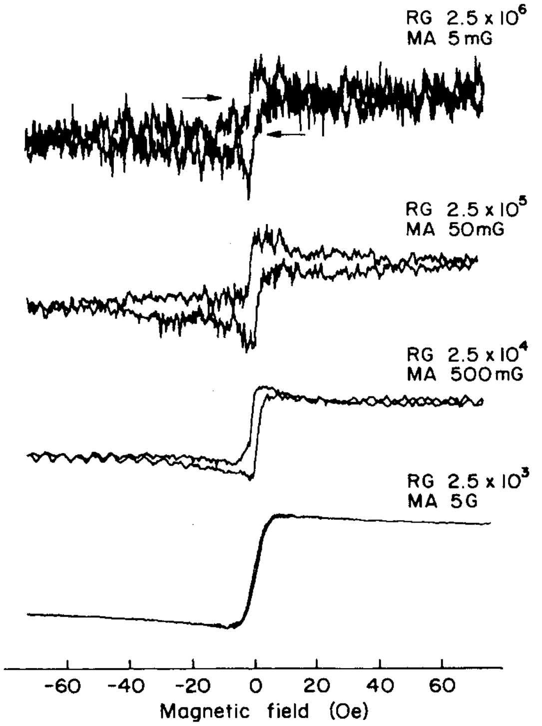

FIG. 12. Microwave signals at 63 K (sample I vacuum annealed for 2 h, $H_0 \parallel c$) with variable modulation amplitude (MA) and receiver gain (RG). The arrows indicate the sense of the magnetic-field sweep.

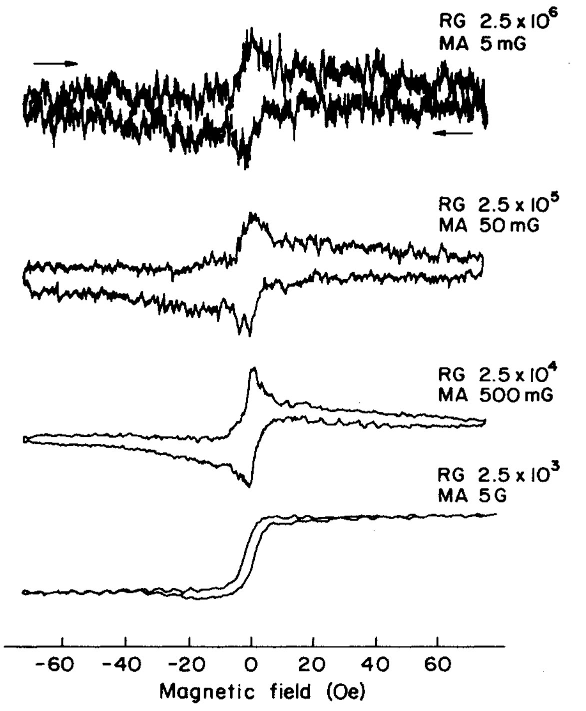

FIG. 13. Microwave signals at 59 K for the same sample as in Fig. 12.

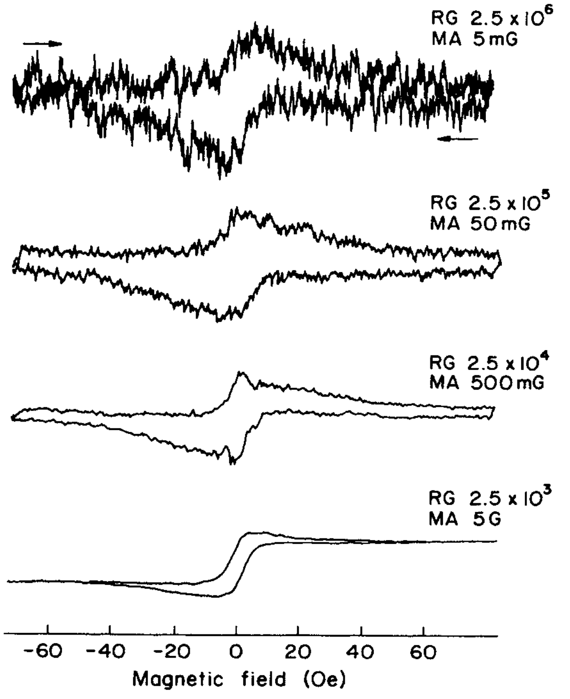

FIG. 14. Microwave signals at 56 K for the same sample as in Figs. 12 and 13.

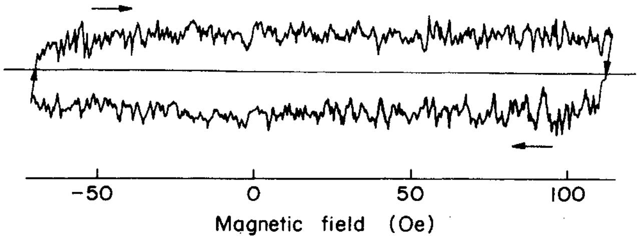

FIG. 15. Microwave looplike signal recorded in forward and reverse field sweeps at 5 K in sample V, and vacuum annealed for 2 h.

linearly with the modulation amplitude (note the change in the receiver gain), until at sufficiently large modulations one starts to induce distortions. The looplike component of the signals in Figs. 12–14 has a different behavior. For a limited field sweep range, it has the form of a rectangular loop, symmetrical with respect to the baseline, i.e., the signal is constant in magnitude, but opposite in sign, for forward and reverse magnetic field sweeps. This component of the signal shows a very sharp response to reversals of the field sweep, i.e., the signal level changes quickly from one trace to the other. A larger modulation amplitude should increase the signal level proportionately, but since it also represents a forward and reverse oscillation of the magnetic field sweep, there occurs a partial averaging of the opposite signal levels of the looplike components. Therefore, this signal component does not grow linearly with the modulation amplitude, and the total signal takes the form as in Figs. 12–14.

At low enough temperatures (usually below 20 K) most of the samples showed just the looplike signal (Fig. 15) with the same characteristics as described above.

VI. DISCUSSION AND CONCLUSIONS

In this paper we have presented the most characteristic features of the microwave signals we have observed in single crystals of $YBa_2Cu_3O_x$. The phenomena are very rich and can help to reveal some structural and magnetic properties of the sample studied. Thus, the annealing study (Sec. III) shows that the oxygen content is not uniform throughout incompletely annealed samples. It seems that the microwave technique could become a useful tool to monitor the improvement of the annealing quality of samples.

In a previous report,[9] we have proposed a possible model for signals A, B, and C. Signal A was related to Josephson junctions at the crystal twin boundaries along (110) planes, following the ideas of Deutscher and Müller.[13] Since the coherence length in the ab plane is an order of magnitude larger than the lattice parameter in this plane, these twin boundaries would act as weak links only within a few degrees below T_c, and then acquire bulk superconducting properties. This evolution could explain the onset and disappearance of signal A as the temperature is decreased. In incompletely oxygen-annealed samples the superconductive state first nucleates in the regions of higher oxygen content, so that twin boundaries within these regions mark the transition by giving rise to signal A. This can be nicely seen in Figs. 2–6.

At lower temperatures the superconductive state extends gradually into the surrounding area of lower oxygen content. We have proposed that in this process clusters of weakly coupled superconducting regions can be formed, giving rise to the superconductive glass state.[15] Signal B has been associated with this state of the system. The annealing study corroborates this model since signal B is greatly suppressed in our most completely oxygen annealed samples where one could expect reasonably uniform oxygen content. On the contrary, signal A is still observed since the twin boundaries do not disappear through the oxygen-annealing process. If vacuum annealing is applied, there occurs a random depletion of oxygen and clusters of weakly coupled superconducting regions can be recovered, giving rise to a strong signal B. The existence of inhomogeneities in $\mathrm{YBa_2Cu_3O}_x$ single crystals has also been established in electron microscopy studies by Sarikaya and Stern.[16] These authors have found significant local variations in the difference between the a and b dimensions of the orthorhombic structure over spatial distances of the order of 1000 Å. Based on these observations, they have also proposed that the superconductive state nucleates first in isolated regions which then grow in size as the temperature is lowered leading to the onset of weak links and the superconductive glassy state.

Two competing models for the magnetic structure of the superconducting state in $\mathrm{YBa_2Cu_3O}_x$ single crystals can be found in the current literature, namely, the conventional flux penetration as in type-II superconductors, and the superconductive glassy state. The Abrikosov vortex structure is supported by the observation (only at 4.2 K) of Bitter patterns.[17] The experiments on magnetization relaxation,[18] and decay of the critical current density[19] have been interpreted as a manifestation of the superconductive glassy state. However, the observed relaxation effects could also be explained by a giant flux creep due to a low pinning energy in systems with a small coherence length.[20] More experiments of that kind should be made to resolve this question definitely. Our present observations suggest that depending on the degree of the oxygen content, one can obtain different magnetic properties which need not reflect the true bulk superconducting state. Therefore, future measurements of that kind should be made with better characterized samples.

The mechanism of microwave absorption is still not clear. For type-II superconductors above H_{c1}, one can have damped motion of fluxons driven by the microwave currents induced on the surface of the sample.[11,12] If there are weak links in the sample, the magnetic field will penetrate through these even below H_{c2}. In that case, however, the critical current of Josephson weak links can be reduced so that a considerable microwave absorption can also occur as a result of the resistance in the Josephson junctions.[21] It is difficult to distinguish this mechanism from the motion of fluxons in the junctions.

We have presented a variety of hysteretic properties associated with the microwave signals in $\mathrm{YBa_2Cu_3O}_x$ single crystals. It is interesting to point out that signal A does not show any hysteresis even for fields which are larger than H_{c1}, estimated to be several hundred oersteds.[7] It can be interpreted either as a lack of pinning in the narrow temperature range below T_c where signal A appears, or as a sign that signal A has a different origin, such as field penetration through weak links. At lower temperatures one can observe hysteretic signals in incompletely oxygen-annealed samples. The width of the hysteresis increases as the temperature is lowered (cf. Figs. 7 and 8), which is an indication of a larger flux trapping. It is interesting to note that the sense of the hysteresis is such that the forward and reverse traces always intersect the baseline before zero field is reached. The same is true for the hysteretic signals in ceramic samples.[4] In the reported microwave measurements without the field modulation,[7] one can notice that the minimum of the absorption occurs before the field sweep reaches zero. This feature has not been pointed out before, but it seems to be intimately related to the diamagnetic properties of the superconducting samples.

We have also presented some rotation experiments (cf. Fig. 10) which need to be discussed. The zero-field rotation has established that the microwave signal depends on the magnetization, while the rotation in a nonzero field has shown that the magnetization is not rigidly bound to the sample. A similar observation was made in direct magnetization measurements on ceramic $\mathrm{YBa_2Cu_3O}_x$, but not on a classical type-II superconductor such as Nb.[22] From the similarity with the results in spin-glass systems,[23] it was concluded that the phenomenon could be indicative of the superconductive glassy state.[22] Again, our findings support the model of superconductive glassy state, but only in incompletely annealed samples.

Finally, we have dealt with the puzzling dependence of the microwave signals on the modulation amplitude (Sec. V). It seems that the total signal in these cases consists of two components, one which grows linearly with the modulation amplitude, and the other which does not. The unusual behavior of the latter is caused by its strong sensitivity to the field sweep reversals, so that the modulation partially averages the forward and reverse traces. It is interesting to mention that the same type of signal, with a prominent looplike component, has been observed also in Nb metal below H_{c1} when the surface of the sample was machined.[6] Light etching of the surface made the signal disappear. Subsequent anodic oxidation recovered the signal but now with a relatively weaker looplike component.[6] In our study of $\mathrm{YBa_2Cu_3O}_x$ single crystals we have usually observed a pure looplike signal at low temperature (cf. Fig. 14). Interestingly, also, polycrystalline $\mathrm{V_3Si}$, synthesized from powders of vanadium and silicon, and having the $A15$ structure, also showed looplike signals below T_c.[5] More extensive studies would have to be done in order to clarify the origin of these signals.

In conclusion, we have shown that the microwave technique is not only a very sensitive method for the detection of superconductivity in new samples, but can also be used to study some properties of the samples. The signals observed in ceramic samples are relatively simple and seem to be dominated by the intergrain couplings. In contrast, the signals observed in single-crystal samples can be diverse and exhibit a rich phenomenology. We were able to relate the microwave observations to the degree of the oxygen anneal of a sample. This could potentially become a useful procedure for testing the achieved quality of samples. We have also established that the microwave absorption depends on the magnetization of the sample, and we have identified a new type of signal which causes an apparent dependence of the total hysteretic signal on the modulation amplitude.[24]

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[24] After this work was completed, there appeared a report by Blazey *et al.* [K. W. Blazey, A. M. Portis, K. A. Müller, and F. H. Holtzberg, Europhys. Lett. **6**, 457 (1988)] on the observation of a series of extremely narrow (2 mOe in width) regularly spaced microwave signals in $YBa_2Cu_3O_x$ single crystals. These were obtained under very different experimental conditions [they include H_1 perpendicular to the *ab* plane (compare with Fig. 1), a very low microwave power level, a very low modulation amplitude, and the temperature significantly below T_c] than those employed in the present work. We have been able to easily reproduce the signals described by Blazey *et al.* in our samples when we employed their experimental conditions, and we confirmed that they are distinctly different from the signals studied in the present work. We observed that the period of the series of lines varies with the rotation of the sample and has a minimum value of 0.7 Oe. Blazey *et al.* have observed a minimum period of 0.2 Oe in a single crystal about 100 μm thick, and a doubling of the period when their crystal cleaved in half. Our observed period on samples about 30 μm thick is consistent with the geometric factor used in the explanation given by Blazey *et al.* We have to point out that these narrow lines do not evolve under different field conditions into the signals *A*, *B*, or *C* studied in this paper. The latter appeared only at higher temperatures where the narrow lines could not be detected. At lower temperatures, the observed narrow lines were smeared out at higher microwave power and/or larger modulation amplitudes so that a signal as shown in Fig. 15 evolved. One may conclude that different experimental regimes are needed to reveal the whole richness of the microwave signals of $YBa_2Cu_3O_x$.