Microwave study of $YBa_2Cu_3O_x$ single crystals

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Absorption of microwaves (9.3 GHz) was measured as a function of an external applied magnetic field in single crystals of $YBa_2Cu_3O_x$. Three distinct types of signals were observed successively as the temperature was lowered from T_c to 2.4 K. They differ in widths, angular dependences, and hysteretic properties. We discuss possible microwave loss mechanisms and a model based on internal Josephson junctions to explain our observation.

Following the discovery of high-T_c superconductors,[1,2] it has been observed by a number of groups that all of these ceramic compounds have remarkable microwave absorption characteristics at low magnetic fields.[3-8] A giant signal is observed to arise just as the temperature is lowered below T_c, and it persists to very low temperatures. It shows hysteresis in forward and reverse magnetic field sweeps. At low temperatures, the exposure of the sample to a moderately high magnetic field results subsequently in a change of the signal amplitude and shape. Also, a rapid change in the magnetic field is accompanied by a large change in the absorption level, and is followed by a relaxation to a new equilibrium value. All of these features have made the microwave measurements an intriguing subject.

In this Brief Report, we present for the first time the results of microwave measurements on single crystals of $YBa_2Cu_3O_x$. The main new feature as compared to the previous results on ceramic samples is the appearance *and disappearance* of *three* distinct signals as the temperature is reduced from T_c to 2.4 K. We discuss the possible loss mechanisms and interpret the extensive temperature dependences, angular distributions, and hysteretic properties of the observed signals in terms of a model which involves internal Josephson junctions in single crystals.

The single crystals of $YBa_2Cu_3O_x$ (typically $2\times1\times0.03$ mm^{-3}) were prepared following the procedure of Schneemeyer *et al.*[9] Microwave measurements were made using an electron spin resonance (ESR) spectrometer and an offset dc magnetic field as described earlier.[5] In this geometry, the microwave magnetic field H_1 is perpendicular to the dc magnetic field H_0.[2-8] Magnetic field modulation at 100 kHz, parallel to H_0, was employed throughout this work. Microwave absorption was then measured with phase-sensitive detection at the fundamental of the modulation frequency to yield the distinct signals reported here. The temperature was measured by a chromel-alumel thermocouple.

Above T_c, the microwave absorption was independent of the magnetic field sweep, i.e., only a baseline could be detected. With a reduction of the temperature below T_c, one could detect the consecutive appearance and disappearance of three distinct signals whose representative forms are shown in Fig. 1, and their respective temperature ranges are shown in Fig. 2. The characteristics of the highest temperature signal, denoted as A, are that it does not exhibit a hysteresis as the magnetic field is swept back and forth, and that it has a very long tail, extending beyond 2 kOe, which means that the actual dc absorption signal is in the form of a wide cusp. Signal B, appearing at a lower temperature, is much narrower than A. Typically, it reaches baseline for fields of about 100 Oe. It is further characterized by the presence of a hysteresis whose width increases as the temperature is lowered. Signal C, appearing at the lowest temperature, is the most unusual. A single sharp line is detected in each forward and backward magnetic field sweep. The width of the hysteresis (the separation of the lines obtained in the two opposite field sweeps) is much larger than the width of the individual lines.

The good temperature separation of the three signals as shown in Fig. 2 occurs only in well-annealed (10 days in

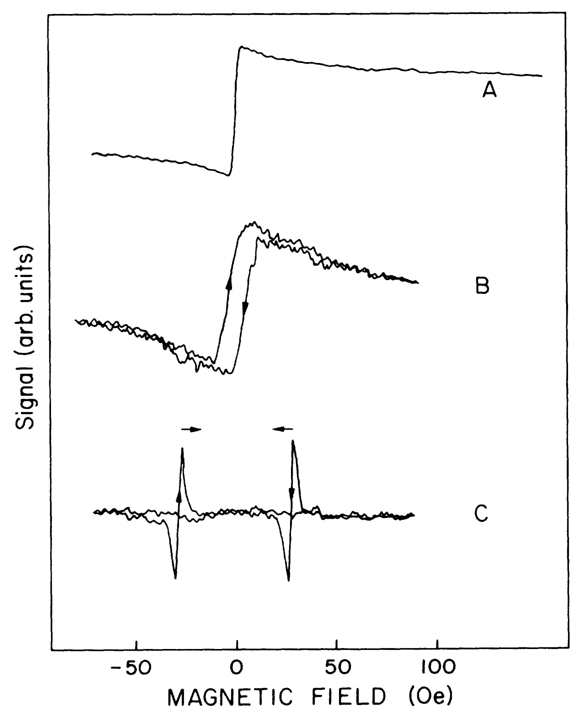

FIG. 1. Representative microwave signals (phase detected at the fundamental of the field modulation) observed in well-oxygen-annealed single crystals of $YBa_2Cu_3O_x$ at temperatures (A) 86 K, (B) 64 K, (C) 36 K and $H_0 \parallel c$.

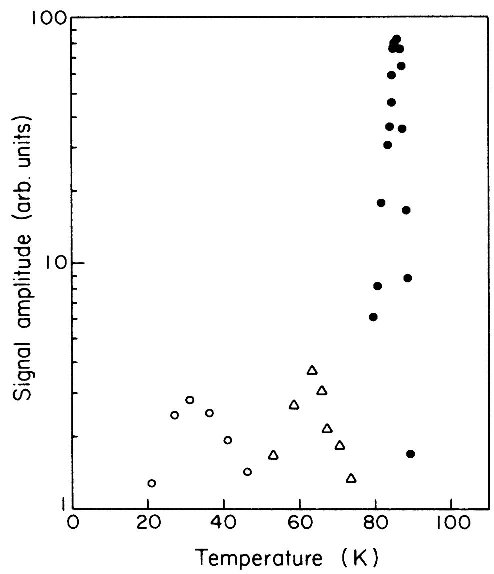

FIG. 2. Temperature dependence of the signals A (●), B (△), and C (○) (cf. Fig. 1) in a well-oxygen-annealed single crystal of $YBa_2Cu_3O_x$ for $H_0 \parallel c$.

oxygen at 490 °C) samples. In less-well-annealed samples, signal A is generally shifted to lower temperatures, so that a composite signal is observed. However, even in these cases, one notices that each component in the signal appears at a given temperature, and vanishes at a lower one. Details of the annealing study will be published elsewhere.

Another interesting feature of these signals is their angular dependence. The sample is rotated around an axis parallel to H_1. The experimental results for signal A are shown in Fig. 3. The peak-to-peak amplitude of the signal has a maximum at $H_0 \parallel c$ (the crystal c axis, perpendicular to the crystal flat face, is defined in Ref. 9) and decreases to zero at $H_0 \perp c$ (with $H_1 \perp c$ in all cases). However, the peak-to-peak width has a small value at $H_0 \parallel c$, and starts to diverge as one approaches $H_0 \perp c$. These angular dependences indicate that the microwave absorption for signal A depends only on the component of H_0 parallel to the c axis. In Fig. 3, we show that the signal amplitude fits well to the cosine law, whereas the peak-to-peak width (as well as the whole line shape) follows the secant law. The angular dependence of signal B is observed to be the same as that of A.

Signal C has particularly interesting properties. Its hysteresis width also grows as the temperature is lowered. Gradually, the signal recorded in each sweep direction starts to broaden, then an unresolved structure appears, and next it splits into a series of partially resolved lines. Figure 4(a) shows an example of this evolution. This splitting is due to an inhomogeneity in the hysteresis width of components of the signal. As the temperature is lowered, the hysteresis width grows for all the components (but faster for the outermost ones), the splitting continues, and the signal gradually approaches a noisy-looking structure. The hysteretic origin of the above splitting can also be verified by recording the signals with decreasing magnetic field scans around zero field. The hysteresis widths gradually shrink, and the splittings are reduced, i.e., the lines are shifted towards zero field. When this procedure is continued to very small field scans around zero, a limiting line shape is traced in both forward and backward field scans without hysteresis. It can be a single unresolved line, but in some favorable cases (of angle, temperature, and sample) a resolved residual structure is observed. An example is shown in Fig. 4(b). This structure cannot be reduced in width by even smaller field scans. Note also that, unlike the hysteretic effect, the residual intrinsic structure yields side lines on both sides of

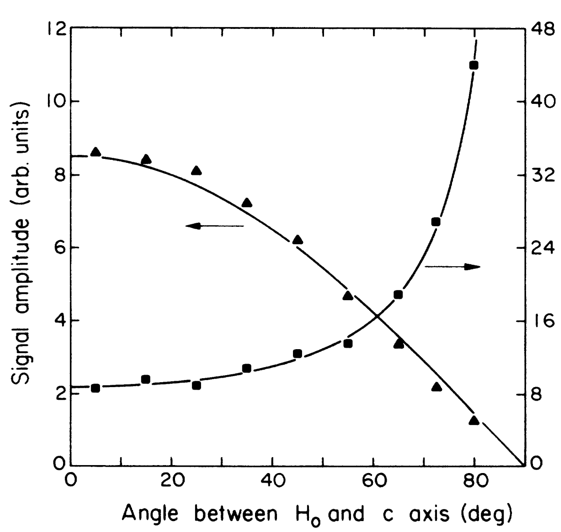

FIG. 3. Angular dependence of the peak-to-peak amplitude (▲) and width (■) of signal A (cf. Fig. 1).

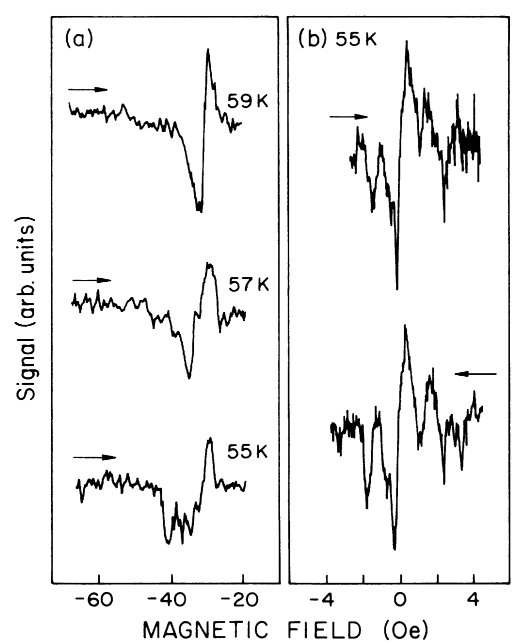

FIG. 4. Signal C is observed at $H_0 \perp c$ in a single crystal of $YBa_2Cu_3O_x$ oxygen annealed for two days. The arrows indicate the sense of the field sweep. (a) One side of signal C at three different temperatures, (b) residual structure at small forward and backward field scans.

zero field. The angular dependence of the amplitude of signal C is very irregular.

Microwave absorption in the presence of a dc magnetic field was measured in type-II superconductors many years ago.[10] The loss mechanism was found to be in the viscous vibrations of fluxons driven by microwave transport currents as a result of the Lorentz force $(1/c)(\mathbf{j}\times\phi_0)$. Thus, this mechanism does not contribute to the microwave absorption when magnetic flux is absent, or when the microwave current is parallel to the applied magnetic field. Recent microwave measurements of single crystals of $YBa_2Cu_3O_x$ in zero magnetic field have shown that the surface resistance drops at temperatures below T_c, but has a nonzero value even at very low temperatures,[11] presumably due to the presence of normal electrons. When a dc magnetic field is applied, as in the present experiment, one may expect that the microwave absorption can be further increased due to the viscous vibrations of fluxons within the superconducting regions, and/or due to the increase of the number of the normal electrons in the vortex cores.

In all our present measurements on single crystals, the signal amplitudes are reduced to the noise level at low temperatures ($\lesssim 10$ K). This is at variance with the results of the previous studies on granular high-T_c superconductors where the signal was almost constant from T_c to very low temperatures.[3-7] Thus, a simple angular averaging of the signals found in single crystals would not yield the observations in granular samples. One may infer that the difference in the signals should have its origin in the coupling between the superconducting grains. A network of Josephson junctions could provide paths for the flux penetration which, in its turn, can give rise to a microwave absorption by viscous vibrations,[10] and/or through a reduced supercurrent screening of the normal electrons in the junctions.

The question remains whether the signals observed in single crystals could be explained assuming that the samples are uniform bulk type-II superconductors. Let us start with the type-A signal. Its angular dependence is in agreement with the viscous flux-flow mechanism. For $H_0 \parallel c$, the microwave induced currents on the sample surface are perpendicular to H_0, and the Lorentz force is maximum. As the sample is rotated to $H_0 \perp c$, the microwave induced currents become parallel to H_0, and the losses due to the viscous flow should vanish. However, the striking feature at the signal A, is its temperature dependence. Its intensity changes by orders of magnitude in a very small temperature interval. Such behavior has not been observed in any previous measurement on strongly or weakly pinned type-II superconductors.[10] Unless one assumes that a dramatic change in the flux viscosity takes place in the present compounds, it may be that a different explanation is needed for the temperature dependence of signal A.

Deutscher and Müller[12] have suggested that very short coherence lengths in $YBa_2Cu_3O_x$ compounds[13] imply that crystal twin boundaries and other defect planes, which are essentially narrow nonsuperconducting regions, can act as internal Josephson junctions. In general, a given boundary will act as a weak link if its thickness is of the order of the coherence length. At temperatures close to T_c, the critical current in a weak link is small, and it increases as the coherence length $\xi(T)$ becomes shorter at lower temperatures. That is[12] $I_0 \propto \Delta_0^2[\xi^2(0)/a\xi(T)]^2 \rightarrow \Delta_0^2[\xi(0)/a]^2$, where Δ_0 is the bulk pair potential. Thus, whether a link remains weak, or becomes strong at temperatures significantly below T_c, depends on the ratio of the low-temperature coherence length $\xi(0)$ to the lattice parameter (e.g., a). With the model of internal Josephson junctions, Deutscher and Müller could explain a number of experimental observations in which the properties of a superconductive glassy state were manifested.[14] Crystal twinning along (110) planes is particularly prominent in $YBa_2Cu_3O_x$.[15] The coherence length in the ab plane was found to be 34 Å at lower temperatures,[13] which is an order of magnitude larger than the lattice parameter. Therefore, one finds that in the temperature range given by $(T_c - T)/T_c \approx [a/\xi_{ab}(0)]^2 \approx 10^{-2}$, the critical Josephson current would increase fast, $I_0 \propto (T - T_c)^2$. For T_c of the order of 10^2 K, this temperature range is of the order of 1 K. Thus, there is enough of an increase in the critical current a few degrees below T_c to transform the twin boundaries along (110) planes from weak lines into strong links. The magnetic flux would thereby be expelled from these boundaries, and the microwave absorption associated with it would vanish. We suggest that this mechanism could serve as a possible explanation for the temperature dependence of signal A.

The form of signal A results from the magnetic field penetration into the junctions. The increasing number of fluxons in the junction gives rise to an increased microwave absorption until the weak link completely breaks due to the field penetration. Alternately, one can say that the penetration of a flux ϕ_j ($\phi_j = 2\lambda d H_0$, λ penetration depth, d junction length) into the junction reduces its supercurrent according to the "diffraction" expression[16] $\sin(\pi\phi_j/\phi_0)/(\pi\phi_j/\phi_0)$, so that the screening of normal electrons is reduced, and the microwave absorption increases, saturating in the limit of large flux penetration. The smaller the size of the junction, the larger are the fields needed to reach this saturation. With a distribution in size of the junctions the individual minima and maxima are smeared out, and the resulting microwave absorption becomes a smooth curve with a single minimum at zero field. A good quantitative measure for the expected shape of the curve is obtained by estimating the fields at which one flux quantum ϕ_0 is passed through a junction. For a penetration depth of the order of 10^3 Å, and a distribution in length of the twin boundaries of $10^3 - 10^4$ Å, the above condition is met for a field range of $10^2 - 10^3$ Oe, which is in good agreement with the observed shape of signal A.

Signals B and C appear at lower temperatures. The same qualitative arguments for a microwave absorption could be applied as in the case of signal A, except that other types of weak links would be required. Due to an inhomogeneous oxygen annealing, some regions within the crystal would become superconducting just below T_c. As the temperature is lowered, the superconducting state extends into the regions of lower oxygen content. This process leads to a formation of loops made by weakly coupled superconducting regions. Signal B could be ascribed to this state of the system. The above model is also supported by our annealing studies. We have found that vacuum annealing depresses signal A, and increases signal B, thus suggesting that initially well-annealed superconducting regions were partially and sporadically reduced in oxygen content. A remarkable property of signal C is that its intensity varies irregularly as the crystal is rotated with respect to H_0, the actual variation being sample dependent. This behavior cannot be reconciled with any homogeneous defect-free property of the sample. We suggest that it is due to weak links formed at a set of planes at different angles. These could be twin boundaries along (001) and/or other defect planes.[15,17] The proposed model is consistent with the occurrence of the inhomogeneity of hysteresis widths shown in Fig. 4(a). The intrinsic structure shown in Fig. 4(b) could be due to the diffraction periodicity associated with the Josephson junctions giving the strongest contribution for that orientation.

Relaxation effects[6] were observed only when hysteretic signals B and/or C were present. They were more pronounced at lower temperatures where the hysteresis was larger. This study, the details of which will be published elsewhere, shows that single crystals indeed exhibit the properties of a superconducting glassy state, as conjectured earlier in the model of intragrain Josephson junctions.[12]

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