Microwave study of YBa$_2$Cu$_3$O$_y$ 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$_2$Cu$_3$O$_y$. 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, it has been observed by a number of groups that all of these ceramic compounds have remarkable microwave absorption characteristics at low magnetic fields. 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$_2$Cu$_3$O$_y$. 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$_2$Cu$_3$O$_y$ (typically 2×1×0.03 mm$^{-3}$) were prepared following the procedure of Schneemeyer et al. Microwave measurements were made using an electron spin resonance (ESR) spectrometer and an offset dc magnetic field as described earlier. In this geometry, the microwave magnetic field $H_1$ is perpendicular to the dc magnetic field $H_0$. 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$_2$Cu$_3$O$_y$ at temperatures (A) 86 K, (B) 64 K, (C) 36 K and H$_{\text{dc}}$.](image-url)
FIG. 2. Temperature dependence of the signals $A$ (●), $B$ (△), and $C$ (○) (cf. Fig. 1) in a well-oxygen-annealed single crystal of YBa$_2$Cu$_3$O$_y$ for $H_{0||c}$.

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 $H_{0||c}$ (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 $H_{0||c}$, 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 hysteric 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 hysteric 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$_2$Cu$_3$O$_y$ 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. 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 \((1/e)(j \times \phi_0)\). 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\(_2\)Cu\(_3\)O\(_x\) 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, presum-
ably 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 (\(< 10\, K\)). This is at variance with the re-
results of the previous studies on granular high-\(T_c\) super-
conductors where the signal was almost constant from \(T_c\)
to very low temperatures. 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 net-
work of Josephson junctions could provide paths for the
flux penetration which, in its turn, can give rise to a
microwave absorption by viscous vibrations, 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 sam-
ple 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
\(H_0 \perp c\), 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. Unless one as-
sumes 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 have suggested that very short
coherence lengths in YBa\(_2\)Cu\(_3\)O\(_x\) compounds 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 bound-
ary 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 tem-
peratures. That is \(I_0 \propto \delta B(\xi^2(0)/a^2)(\delta \xi(0)/a)^2\),
where \(\delta B\) 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 para-
ter (e.g., \(a\)). With the model of internal Josephson junc-
tions, Deutscher and Müller could explain a number of ex-
perimental observations in which the properties of a super-
conducting glassy state were manifest.\(^{14}\) Crystal
 twinning along (110) planes is particularly prominent in
YBa\(_2\)Cu\(_3\)O\(_y\).\(^{15}\) The coherence length in the \(ab\) plane was
found to be 34 \AA\ 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
\((T_c - T)/T_c = [\alpha/\xi(0)]^2 \approx 10^{-2}\), the critical
Josephson current would increase fast, \(I_0 \propto (T_c - T)^2\).
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 ex-
peled from these boundaries, and the microwave absorp-
tion 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 micro-
wave absorption until the weak link completely breaks
due to the field penetration. Alternately, one can say that
the penetration of a flux \(\phi_j (\phi_j = 2\pi \lambda d H_0, \lambda
penetration depth, \(\lambda\) junction length) into the junction reduces its
supercurrent according to the "diffraction" expression
\(\sin(\pi \phi_j/\phi_0)/(\pi \phi_j/\phi_0)\), 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 \(\phi_0\) is passed through a junction. For
a penetration depth of the order of 10\(^3\) A, and a distribution
in length of the twin boundaries of 10\(^3\) - 10\(^4\) A, 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 support-
ed 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.\textsuperscript{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\textsuperscript{8} 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.\textsuperscript{12}

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\textsuperscript{17}M. Hervieu \textit{et al.}, Europhys. Lett. 4, 205 (1987); B. Demenges \textit{et al.}, \textit{ibid.} 4, 211 (1987).