Microwave absorption across T_c : Determination of the angular dependence $H_{c2}(\theta)/H_{c2}$

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It is shown that measuring microwave absorption in high- T_c superconductors at constant and very low magnetic fields, using magnetic-field modulation, is, under some conditions, equivalent to temperature modulation when sweeping the temperature across T_c . Using an ESR spectrometer, the derivative of microwave absorption is measured close to T_c . This allows a determination of the relative angular variation of dH_{c2}/dT at $T = T_c$ in single crystals of Y-Ba-Cu-O. The data fit the Ginzburg-Landau theory on the relative angular variation of H_{c2} . The ratio $(dH_{c2}/dT)_{T=T_c}$ parallel and perpendicular to the Cu planes was found to be 2.7 and 5.3 for two Y-Ba-Cu-O single crystals with $T_c = 89$ and 86 K, respectively. These values obtained at 10¹⁰ Hz are close to the values obtained by conventional dc methods.

INTRODUCTION

Microwave absorption has been used to investigate the properties of superconducting substances. Various techniques have been introduced where either only microwave power has been applied¹ or a combination of microwave power and magnetic fields.²⁻⁴ In the first method, the microwave impedance is measured. One can then derive the penetration depth $\lambda(f, T)$ where f is the frequency, which is important in evaluating the superconductivity theories.¹ The microwave impedance, as a function of temperature, is usually obtained from measurements of changes of the quality factor Q and the shift of the resonance frequency of a microwave cavity with and without the superconducting samples.¹ An alternative method would be to use temperature modulation of the sample in the cavity and simultaneously sweep the temperature leading to derivative of the microwave resistance, dR(T)/dT curve. Recently it was shown that strong signals are obtained when sweeping the temperature through the superconducting transition temperature T_c using a conventional electron spin resonance (ESR) spectrometer with magnetic-field modulation.^{5,6} These signals resemble the temperature derivative of the microwave surface resistance. Indeed, as is shown below, magnetic-field modulation in high- T_c superconductors with high- H_{c2} values is approximately equivalent to temperature modulation when crossing the transition temperature T_c . In this way the derivative of the resistivity, with respect to temperature, is obtained.

In this work we show how one can use this effect to measure the relative angular variation of dH_{c2}/dT in high- H_{c2} superconducting single crystals. This method allows a characterization of the superconducting properties in single crystals and thin films. In the following we discuss the theoretical aspects, followed by the experimental results and their comparison with theory.

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THEORETICAL CONSIDERATIONS

We first present a qualitative argument that shows how magnetic modulation at $T = T_c$ can be presented as temperature modulation.⁷ The experiment is carried out by measuring the reflected power from a microwave cavity that contains the superconducting material when sweeping the temperature from above to below T_c . When crossing the $H_{c2}(T)$ curve at $T = T_c$ at a low magnetic field H_0 , using magnetic modulation $H_m = H_1 \sin \omega t$ $(H_0 > H_1)$ a positive value of H_m will bring the system to the normal state while a negative value of H_m will drive the system into the superconducting state. Hence the Qfactor of the cavity will change due to the change of the microwave surface resistance when going from the normal to the superconducting state. This magnetic modulation at $T = T_c$ has the same effect as temperature modulation $T_m = T_1 \sin \omega t$ where positive and negative values of T_m will drive the system into the normal and the superconducting state, respectively. The change into the superconducting state and therefore the microwave resistivity will be inversely proportional to the slope of the H_{c2} curve versus temperature, hence the intensity of the signal will also be inversely proportional to dH_{c2}/dT .

We now proceed to a quantitative description. Our central simplifying assumption is that the microwave surface resistance varies as a function of the temperature Tand the field H, similar to the dc resistance R(T,H). This crude approximation is not expected to hold in high fields, where the viscous motion of vortex lines can cause a frequency dependence of the resistivity. But in the situation of interest—low bias fields and close to T_c —the major component of the surface resistance originates from normal-state inhomogeneities and from dissipation across Josephson junctions ("weak links"), a basically dc effect. A typical plot of R(T,H) is given in Fig. 1 (see,



FIG. 1. Schematic presentation of microwave surface resistivity R as a function of temperature and magnetic field, where it is assumed to have a functional behavior similar to the dc resistivity. It shows that magnetic-field modulation, when sweeping the temperature, will result in a peak in the demodulated microwave signal slightly below T_c .

e.g., Refs. 8 and 9). Figure 1 also shows how the bias field H_0 (≈ 30 Oe in our experiment) and the modulation ΔH (≈ 1 Oe) give rise to a signal that depends on $\Delta r \approx (\partial R / \partial H)_{H_0} \Delta H$ in isothermal conditions. If the temperature is varied while H_0 and ΔH are held constant, the demodulated ESR signal becomes maximum somewhat below T_c (Fig. 1, right part).

Reversibility is generally observed in a $\sim 5K$ window below T_c for high-temperature superconductors.¹⁰⁻¹³ The resistance R(H,T) obeys then a state equation. Using the identity

$$(\partial R / \partial H)_T (\partial H / \partial T)_R (\partial T / \partial r)_H = -1$$
,

we can then relate the ESR signal I to the resistance derivative and to the slope of the upper critical field H_{c2} :

$$I \propto \Delta R = \Delta H \left[\frac{\partial R}{\partial T} \right]_{H_0} / \left[\frac{\partial H_{c2}}{\partial T} \right] . \tag{1}$$

We have made use of the fact that the intersection of the R(H,T) surface with the R=const plane (e.g., $R=R_n/2$, where R_n is the normal-state resistance;see Fig. 1) is a common, albeit not rigorous, definition of the $H_{c2}(T)$ curve.¹⁴ Introducing further the Werthamer-Helfand-Hohenberg (WHH) estimate¹⁵ $H_{c2}(T=0) = 0.7T_c(dH_{c2}/dT)_{T_c}$, we find that the spectrometer signal is proportional to the quantity

 $(\partial R / \partial T)_{H_0} [T_c / H_{c2}(0)] \Delta H$.

It provides, therefore, a measure of the slope of the resistive transition as a function of the temperature, as suggested by the experimentally observed shape. We should mention, however, that the proportionality between I and $(\partial R / \partial T)_{H_0}$ is approximate since $(\partial H / \partial T)_R$ is not constant, but varies somewhat with R. In other words, $(\partial H_{c2} / \partial T)_{T_c}$ depends on the resistance criterion that defines the transition temperature. This problem is of lesser importance if one restricts the analysis to the region where the signal I is strong, thus avoiding large variations of $(\partial H / \partial T)_R$. In particular, the critical field H_{c2} , defined by the criterion of the steepest point of the resistance transition, determines the peak value of I in Eq. (1).

We observe that the signal intensity I depends on the angle between H_0 (and ΔH) and the *c* axis. For the small bias fields used in these experiments, the quantity $(\partial R / \partial T)_{H_0}$ is close to isotropic zero field value (see, however, Ref. 9 for relevant data at higher fields). The variation of the demagnetizing factor as a function of the angle may be neglected because we map the transition into the mixed state of an extreme type-II superconductor. The anisotropy of the ESR absorption signal is therefore a consequence of the anisotropy of the critical field. The latter is conveniently described in uniaxial superconductor.¹⁶

$$H_{c2}(\theta) = H_{c2}(n) [m_3 / m(\theta)]^{1/2}$$

= $H_{c2}(n) [\cos^2 \theta + m_1 / m_3 \sin^2 \theta]^{-1/2},$ (2)

where $m_1 = m_{xx}$, $m_2 = m_{yy}$, $m_3 = m_{zz}$, $H_{c2}(n)$, and $H_{c2}(p)$,

$$I(\theta)/I(n) = \{\cos^2\theta + [H_{c2}(n)/H_{c2}(p)]^2 \sin^2\theta\}^{1/2}.$$
 (3)

Thus, measuring the ratio of the microwave absorption intensities normal [I(n)] and parallel [I(p)] to the CuO planes in a single crystal should give the ratio of the critical fields in these directions (or, more precisely, of their slopes at T_c). Furthermore, the angular dependence of the critical field should follow Eq. (3).

EXPERIMENTAL AND RESULTS

Microwave absorption near T_c in ceramics was demonstrated in earlier publications.^{5,6} Here we will discuss only YBa₂Cu₃O₆+x single crystals; oriented thin films are planned to be discussed elsewhere.

Y-Ba-Cu-O single crystals were grown by the flux method.¹⁷ Their c axis was perpendicular to the crystal plane. A Varian E-line spectrometer was used with a homemade TE_{102} cavity. The modulation coils were attached to the magnet poles. Rotating the magnet will turn the dc as well as the modulating field keeping all other parameters constant. The crystal was attached to the side wall of the cavity and therefore H_{rf} was always perpendicular to the magnetic field. The sweep of the



FIG. 2. Peak microwave absorption at the transition as a function of θ , the angle between the *c* axis and the magnetic field, for two Y-Ba-Cu-O crystals. The solid line is a fit to Eq. (3) derived from the Ginzburg-Landau theory. The shape of the ESR signal as a function of temperature is shown for two representative cases; their height is proportional to their intensities.

temperature was always from above to below T_c .

Figure 2 shows the variation of the intensity of the signal as a function of θ , the angle between the normal to the Cu planes (i.e., the direction between the c axis and the magnetic field) of two different single crystals. The intensity was taken at the maximum of the recorded temperature dependence for each angle. Samples with a well-defined single peak were selected for this study. The microwave absorption signal for the two extreme directions of the higher- T_c sample are shown as insets. The width at half intensity is less than 1.5 K. The shape of the signal did not vary with the angle. This was true for all measured samples. This important observation supports the implicit assumption that the variation of the signal as a function of the angle is not due to an anisotropy of the pinning in the flux flow regime, which would broaden the transition. Our accuracy in measuring the temperature was not enough to extract the functional dependence of the peak as a function of the angle. The solid lines of Fig. 2 are presented by Eq. (3) derived from the Ginzburg-Landau theory. They follow the experimental results quite closely. The fit was obtained assuming $H_{c2}(p)/H_{c2}(n)=5.3$ and 2.7 for the samples with $T_c = 86$ and 89, respectively.

DISCUSSION

The novel feature of this method is that magnetic modulation at T_c corresponds to temperature modulation whose amplitude is $\Delta T = \Delta H / (dH_{c2}/dT)_{T=T_c}$. Keeping all variables, except the orientation of the magnetic field, constant, and as the intensity is proportional to ΔT , we were able to measure the anisotropy of the derivative



FIG. 3. Microwave absorption around T_c for another crystal $2 \times 2 \times 0.3$ mm³. The transition width for this single crystal as determined by ac susceptibility is 2 K. The solid line is a best fit assuming that the crystal contains a few regions whose transition is Gaussian with $T_c = 86.9$, 86.2, 85.5, and 83.6 and ΔT , the full width at half width, is 1.24, 0.39, 1.13, and 0.6 K, respectively.

 (dH_{c2}/dT) at the transition temperature T_c .

The analysis of the results is based on the assumption that the microwave losses are proportional to the dc resistance and that the system behaves reversibly. With this approximation we fit our results to the Ginzburg-Landau theory on the relative variation of H_{c2} as a function of θ . The only parameter used to fit the data is

$$S = I(n)/I(p) = [dH_{c2}(p)/dT]/[dH_{c2}(n)/dT]$$

at $T \approx T_c$. The fit in Fig. 2 is good though it shows some deviations. This is not surprising in view of our approximations. In the two crystals measured we obtain S at $T = T_c$ equal to 5.3 and 2.7 for T_c 86 and 89 K, respectively, where T_c had been chosen as the temperature of the peak of the intensity. These results may be compared with other determinations. Bauhofer *et al.*¹⁸ have measured critical fields determined by the onset of flux motion at approximately 10 MHz in an untwinned single crystal of YBa₂Cu₃O_{6+x}. Depending on the way tangents are drawn to their H_{c2} versus T plots, anisotropy ratios between 3 and 5 are found for x = 0.9. They also found a large increase of S with the decrease of the oxygen content. Thus we expect that S will increase with decreasing T_c (as T_c decreases when x is reduced) in agreement with our experimental results cited above. Other

measurements on the anisotropy of microwave absorption at lower values of x are planned to check this point. It would be extremely important to compare the angular variation of dH_{c2}/dT obtained from the microwave absorption measurements with results obtained by the dc methods, using the same sample. As it was pointed out by Malozemoff et al.¹⁹, different techniques give different values at low magnetic fields. Such a comparison may be significant with regard to the frequency response of the H_{c2} line.

Welp et al.²⁰ have extracted the angular anisotropy in a Y-Ba-Cu-O sample $T_c = 92.5$ from the angular variation of T_c at 5T, and also found an agreement with the Ginzburg-Landau theory, with S = 5.

Finally, it should be mentioned that microwave absorption signals in other single crystals may show such structures as seen in Fig. 3 and have been reported earlier.⁶ We believe that the origin of this structure is due to different regions in the single crystal that exhibit slightly different T_c 's and transition temperature width, though the x-ray measurements show single crystal structure.

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