Microwave absorption of aligned crystalline grains of $YBa_2Cu_3O_{7-x}$

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We investigate here the anisotropic magnetic and electric properties of aligned crystalline grains of $YBa_2Cu_3O_{7-x}$. The microwave absorption is indeed consistent with the expected temperature behaviors of the resistivity in the normal state along the *c* axis and in the *a-b* plane. In the superconducting state, both the absorptions in pure rf electric and magnetic fields are high compared to the conventional superconductors; for the magnetic case activation energies are deduced and the dispersion is related to the diamagnetic susceptibility. These results are discussed qualitatively in relation to similar data obtained on single crystals and ceramic samples.

INTRODUCTION

It is well known that anisotropy is a characteristic feature of the 90-K ceramic superconductors and several experimental studies are found in the literature to confirm this assertion. These are, however, performed on small volume single crystals for which the transition temperature is often less than 92 K. Among these, transport measurements[1] are especially difficult to realize because of the need for good electrical contacts on very tiny crystals. Nevertheless, magnetization and transport measurements have been performed on single crystals and the anisotropic character of the electrical and magnetic properties confirmed. Microwave techniques[2] may also be used for the same purpose but many crystals have to be oriented to get enough absorption sensitivity. It should, however, be interesting and desirable to obtain the same type of information for crystals produced by the standard processing route.

SAMPLE AND EXPERIMENT

When a crystal possessing anisotropy in its paramagnetic susceptibility is placed in a magnetic field, it will tend to align so that the direction having the greatest magnetic susceptibility lies along the field. This property has been used with success to produce uniaxially aligned small single crystals of $YBa_2Cu_3O_{7-x}$ (Ref. 3); with available magnetic fields and a suitably chosen medium, the alignment of the tiny crystals can be rapidly established even if the torques involved are small. The $YBa_2Cu_3O_{7-x}$ powder was obtained from samples prepared using the standard ceramic powder processing. The roughly spherical single-crystal grains of the resulting powder were 3–6 μm in diameter and the original T_c, according to the magnetic susceptibility curve, was around 90 K. A permanently aligned sample was produced by mixing the powder (15% in volume) with Stycast 1266 commercial epoxy resin, then curing the mixture for a few hours in a magnetic field of 7.85 T at room temperature. It is expected that uniaxial ordering would be produced with the *c* axis lying along the field direction and with the *a* and *b* axes having random orientation in the plane perpendicular to the field. The alignment of the sample relative to the *c* axis was better than $\pm 3°$ as confirmed by the x-ray rocking curve.

Although magnetization may be obtained from such samples, the superconductor content is too low to allow dc resistivity measurements. We will thus rely on a microwave absorption technique in order to get the electrical and magnetic properties of our aligned sample. A cavity perturbation method[4] is used to measure the microwave absorption in pure rf electric and magnetic fields at 16.8 GHz. The absorption is measured as a function of temperature (4–300 K) and external magnetic field (0–8 T) applied perpendicular to the microwave field. According to the technique, the variation of the quality factor of the cavity $\Delta(1/2Q)$ and the shift of its resonance frequency δ, after sample insertion at the location of the maximum microwave electric or magnetic field (TE_{102} resonance mode), are registered at each temperature. Parallelepiped samples ($5\times1\times0.5$ mm^3) were cut from the aligned one with the *c* axis of the grains directed parallel or perpendicular to the largest edge; a similar sample with 100% epoxy resin was also investigated.

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RESULTS AND DISCUSSION

For the epoxy resin the technical bulletin of the manufacturer indicates a dielectric constant ϵ_1 around 3 and a dissipation factor ϵ_2 of 0.02 in the megahertz range. At microwave frequencies (16.8 GHz), ϵ_1 is still around 3 but the dissipation factor is higher ~ 0.1. As measured by our technique (Fig. 1), both parameters are temperature dependent over the 4–300-K range, especially for $T > 100$ K. Such dielectric effects of the epoxy should be considered for the interpretation of the microwave absorption in pure rf electric field of the composite samples. Over the same frequency range, microwave magnetic field effects have not been observed.

We present in Fig. 2 the relative microwave absorption $\Delta(1/2Q)$ in pure rf electric field of composite samples having a similar geometry but a different orientation of the c axis relative to the microwave field. Both sets of data show a high-temperature tail which is dominated by the dielectric absorption of the epoxy matrix, and a clear signature of the superconducting phase transition around 90 K. The data concerning the resonance frequency shift are also dominated by the epoxy, but no particular structure is seen around T_c, and this is why the data are not shown here.

The discussion of the data presented in Fig. 2 could be facilitated if one could substract the dielectric contribution of the matrix to the absorption. The effective-medium theory[5,6] provides a procedure for that purpose when the radius of the inclusions is smaller than the skin depth; even if this approximation is valid here, the procedure has not yielded reliable parameters. Besides the effective-medium theory, we have also tried the very simple direct addition of the absorptions $\Delta(1/Q)$, matrix, and superconductor, but unreasonable results were also obtained for the normal-state resistivities. Since no quantitative data analysis is possible to extract these parameters from the electric component of the absorption, we will thus present a qualitative explanation.

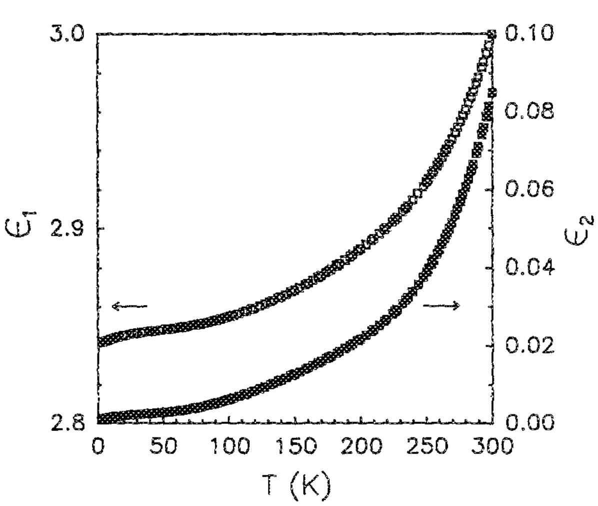

FIG. 1. Complex dielectric function $\epsilon_1 - J\epsilon_2$ of Stycast 1266 resin at 16.8 GHz as a function of temperature. ϵ_1: dielectric constant; ϵ_2: dissipation factor.

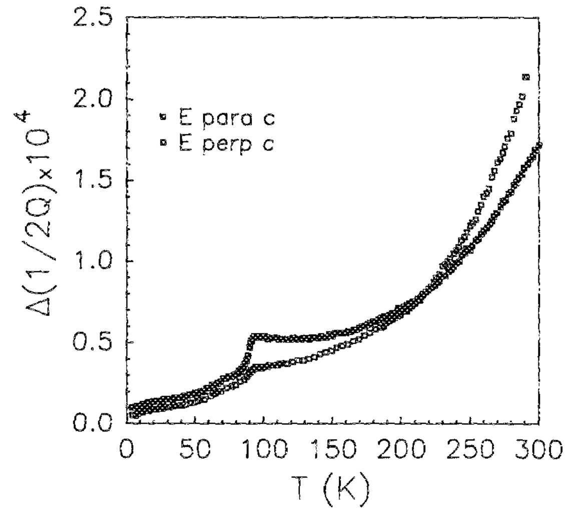

FIG. 2. Relative microwave absorption $\Delta(1/2Q)$ in a pure rf electric field as a function of temperature for two field orientations.

Below 100 K we notice in Fig. 1 that the epoxy's absorption becomes negligible; it thus seems possible over this temperature range to discuss only the $YBa_2Cu_3O_{7-x}$ contribution. Above 100 K the epoxy dominates and it is not possible to isolate the superconductor contribution. For a small conducting sphere exposed to a homogeneous rf electric field, the relative losses are given by the quasi-static equation[7] if the skin depth and the vacuum wavelength are larger than the sphere radius,

$$\Delta(1/2Q) = \omega\alpha\rho\epsilon_0/N^2. \quad (1)$$

It is assumed here that the dielectric losses are much smaller than the ohmic ones. ω, α, ρ, ϵ_0, and N are, respectively, the angular frequency, the cavity filling factor, the resistivity, the free-space permittivity, and the depolarization factor (1/3 for a sphere). The losses are then directly proportional to the sample resistivity.

When the electric field is perpendicular to c, the microwave currents are in the a-b plane and the absorption should be the image of ρ_{ab}, the resistivity in the superconducting a-b planes. For $T < 100$ K, a small variation of the absorption is seen (in Fig. 2) around T_c and a quasi-linear increase with temperature is observed; this is consistent with the expected temperature behavior of ρ_{ab} [Eq. (1)]. When the field is parallel to c, the absorption should rather be the image of ρ_c, the resistivity along the c axis. The absorption shows indeed a maximum at T_c in agreement with ρ_c increasing when the temperature is lowered.[1] Here the trend is a higher absorption along the c axis as compared to the perpendicular direction, an observation consistent with $\rho_c > \rho_{ab}$. Absolute values are difficult to obtain from Eq. (1) since different N must be used for the c and ab directions. If the grains have more or less the shape of an elongated ellipsoid (long axis in the ab plane), the anisotropy ratio ρ_c/ρ_{ab} at 90 K should be higher than what is suggested in Fig. 2; the correction factor $(N_c/N_{ab})^2$ is expected to be much higher than 1 since $N_c > N_{ab}$.

In the superconducting state ($T < T_c$) the absorption for both directions decreases with decreasing temperature; rapidly at first and then more slowly. Compared to the BCS superconductors, the absorption is important below T_c since the matrix losses are negligibly small. This level of absorption is comparable to the one observed in the ceramic compounds[4] where intergrain weak links are important; here the absorption could partly be due to intragrain weak links or to a residual phase.

The data concerning the microwave absorption in a pure rf magnetic field are presented in Figs. 3(a) and 3(b) for both field orientations. As the epoxy matrix is nonmagnetic over this temperature range, they should, in principle, be easier to understand. When the field is oriented along the c axis, Fig. 3(a), the cavity frequency shift δ (dispersion) is slightly negative and constant for $T > 90$ K. At 90 K, the onset of the superconducting transition, the shift drops rapidly and saturates below 40 K. The losses $\Delta(1/2Q)$ show a very sharp maximum at 90 K with different temperature behaviors for $T > T_c$ and $T < T_c$. When the field is rather aligned in the a-b plane, Fig. 3(b), the absorption is much smaller; it shows a maximum around 80 K and the data practically saturate for $T > T_c$. The frequency shift is, however, similar to the one observed for the parallel orientation; the superconducting onset is still at 90 K with a slightly positive value for $T > T_c$ but always negative below.

We know from microwave experiments on magnetic structures[8] that the microwave absorption and dispersion are related to the complex susceptibility function. For conductors however, eddy current losses are also to be considered since they will dominate the absorption. For conducting spheres in a pure rf magnetic field, the losses have been calculated[7,9] following the same assumptions stated for the pure electric case: the radius of the sphere is much smaller than the vacuum wavelength and the skin depth is large compared to the radius. As it has been said before, the skin depth is larger than the radius of the grains according to the normal-state resistivity values found in the literature[1] and we will thus consider all these assumptions to be valid in our experiments. If one neglects the magnetic losses, the measured absorption $\Delta(1/2Q)$ are then inversely proportional to the resistivity ρ (it was proportional to ρ for the electric case); the following equation is indeed obtained for the particular geometry of resonant cavity used in the experiment:

$$\Delta(1/2Q) = \alpha R^2 \omega \mu_0 / 20\rho. \quad (2)$$

R is the average radius of the grains and μ_0 the free-space permeability. The frequency shift will also be dependent on the conductivity but as the measured shift for $T > T_c$ is small compared to the error, these data will not be discussed here.

When the field is along the c axis, the induced currents are in the a-b plane and the losses should be proportional to $1/\rho_{ab}$. In Fig. 4, we present the normal-state resistivity data as obtained with Eq. (2) for the parallel configuration. A linear relation $\rho_{ab} = 40\,\mu\Omega\text{ cm} + 0.282\,\mu\Omega\text{ cm}/k\,T$ fits the data for $T > T_c$. The constant term and the slope are both smaller than the values found in the literature[1] and this could be due to the fact that we have approximated the grains by spheres (1/3 depolarization factor) with average radius R. If more appropriate platelike structures are used, higher values are likely to be found. When the field is rather perpendicular to the c axis; the currents are in the a-b plane and also along the c axis; the losses should then be proportional to an effective conductivity. The sensitivity however is not very good because of a weak absorption, but the data are practically temperature independent, a situation which might be consistent with a mingling of ρ_{ab} and ρ_c (ρ_c usually showing a maximum at T_c). So the absorption data are indeed the image of the normal-state resistivities when eddy current losses are dominating.

In the superconducting state the origin of the absorption

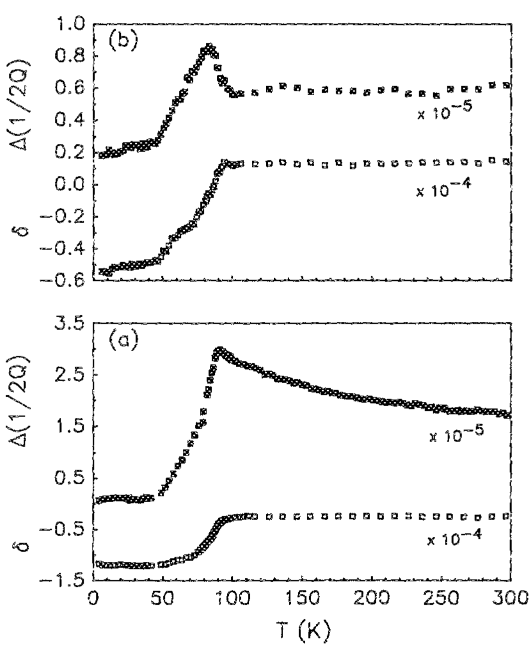

FIG. 3. Relative microwave absorption $\Delta(1/2Q)$ (■) and frequency shift δ (□) in pure rf magnetic field as a function of temperature for two field orientations: (a) H parallel to c and (b) H perpendicular to c.

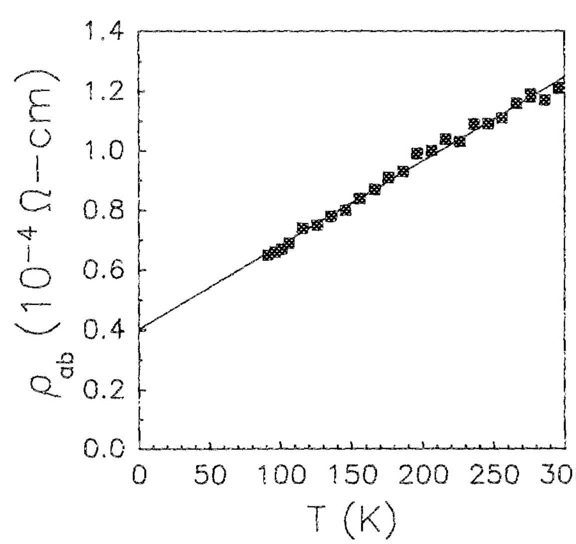

FIG. 4. Resistivity in the a-b plane as deduced from the microwave absorption obtained with H_{rf} parallel to c. A linear regression (continuous line) gives $\rho_{ab} = 40\,\mu\Omega\text{ cm} + 0.282\,\mu\Omega\text{ cm}/k\,T$.

is not so clear. The frequency shift data of Figs. 3(a) and 3(b) show the usual behavior of the real part of the susceptibility χ'; negative values are consistent with the diamagnetic character of the superconducting state and the temperature dependence is similar to the dc susceptibility obtained on a powder sample. The frequency shift may also be related to the surface reactance of the grains or, in other words, to the London penetration depth; however, uncertain geometrical factors hinder the extraction of absolute values and no well-characterized temperature behavior is observed.

On the other hand, the losses are related to the imaginary part χ'' which generally shows a peak[10] below T_c for these high-T_c superconductors. In ceramic samples at low frequencies this peak has been related to hysteresis loss, in part because of sample inhomogeneity and coupling between superconducting inclusions. Such a hysteresis peak is not seen in the parallel configuration; below T_c the decrease is rather progressive and these losses can be related to the surface resistance R_s which, for the BCS case, generally shows a $1/T$ behavior in relation to the superconducting gap. Here a single-gap value does not emerge; our data can be fitted reasonably well by two $1/T$ exponential terms. The first activation energy is highly dependent on an external magnetic field: 310, 253, 182, and 132 K ($\pm$ 10 K) for, respectively, $H = 0$, 1.53, 3.54, and 8.59 T; the second activation energy 3 ± 1 K is practically field independent. In the perpendicular configuration, the absorption below T_c is lower, but a peak is seen around 80 K which may be related to a hysteresis loss. Below 80 K, activation energies of 202 ± 10 K and 3 ± 1 K are deduced in the zero field. Since the absorption is small, no external field dependence study has been done. It thus seems that, even if the real part χ' has the same temperature dependence for both configurations (London penetration depth), the loss data are different below T_c. Evidence is present for the coexistence of hysteretic and resistive losses. Even if two activation energies are measured, it does not mean that two superconducting gaps are to be considered; we rather believe that they are an extrinsic property of the sample as one may find in the literature various temperature behaviors for the microwave surface resistance in the superconducting state.

CONCLUSION

In conclusion our microwave data confirm the anisotropic character of $YBa_2Cu_3O_{7-x}$ aligned crystalline grains in the normal state. Both microwave absorptions in pure rf electric and magnetic fields yield temperature dependencies in agreement with the measured anisotropy on single crystals. Unluckily, the absorption in the electric field is dominated by the epoxy losses which could not be isolated from the superconducting material ones. For $T < T_c$, the data confirm the relatively important absorption in the superconducting state which may be due to other phases or superconducting weak links. The absorption in the magnetic field, however, is solely due to the superconducting material and the anisotropic character has thus been confirmed for $T > T_c$; the obtained temperature dependence of ρ_{ab} is similar to what is found by dc measurements on single crystals. Below T_c the absorption is still high and activation energies are deduced. Hysteresis and resistive losses are likely to explain the observed absorption and no single value of a gap could be extracted.

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