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MICROWAVE ABSORPTION AND JOSEPHSON JUNCTIONS IN SUPERCONDUCTING

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We report the results of microwave absorption measurements from 4 K to T_c in magnetic fields up to 12 kG. The observed strong, magnetic field dependent absorption provides an extremely sensitive and convenient (sample geometry independent) method of characterizing the new high-T_c superconductors and of detecting the possibly higher T_c phases in new samples. Results are also presented on a microwave radiation induced dc voltage across the superconducting samples. This voltage is also magnetic field dependent and it peaks at zero magnetic field. We propose a model for the low-field absorption and a possible relation of this absorption to the microwave induced dc voltage.

I. INTRODUCTION

The recent discoveries [1—4] of superconductivity at record high temperatures have opened a new and exciting chapter in the field of superconductivity. Superconducting transition temperatures of 125 K and perhaps higher promise technical application of superconductivity on an unprecedented scale. Researchers from all over the world are putting immense efforts to understand the mechanism of the high-T_c superconductivity and the behaviour of the new superconductors through experimental and theoretical investigations.

One of the most interesting properties of the new high-T_c superconductors is a strong, magnetic field dependent, non-resonant microwave and radio frequency absorption first reported for these materials by us [5—7]. We have primarily studied the Y-Ba-Cu-O system, but the absorption is present in other high-T_c compounds as well. Recently, several groups reported absorption at very low magnetic fields and associated it mainly with the existence of the Josephson junctions [8—14]. According to Portis et al. [15] the microwave absorption in high-T_c oxide superconductors arises from the microwave conductivity loss through the dissipation from the fluxon motion driven by microwave induced currents.

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Here, we present the results of our microwave absorption measurements on different samples having the same nominal $YBa_2Cu_3O_7$ composition but differing in preparation. We have also measured a microwave induced dc voltage across the samples placed in a microwave cavity. We propose a model based on long Josephson junctions for the non-resonant microwave absorption in high-T_c superconductors and a possible relationship between this absorption and the induced voltage.

II. EXPERIMENTAL

The superconducting $YBa_2Cu_3O_7$ ceramic samples were prepared by the solid state reaction of $BaCO_3$ (reagent grade), Y_2O_3 (99.999%), and CuO (puratronic grade) [16]. All powders were mixed and ground for 24 hours with ethanol and alumina. Mixed powders were dried and fired in an alumina crucible at 940 °C in air for 50 hours. Subsequently, calcined powder was milled for 24 hours and a binder added. Pellets with a diameter of 1.5 cm and a thickness of 0.3 cm were pressed from the powder-binder mixture. The pellets were fired at 970 °C for 24 hours and cooled at a rate of 50 °C/hour. The binder was completely removed during the sintering. The pellets were then annealed in oxygen and then cooled at a rate of 30 °C/hour. We have found that the samples are sensitive to the conditions of annealing in oxygen and prepared, under control, two groups of samples, A and B. The obtained samples exhibited an orthorhombic single phase as confirmed by X-ray powder diffraction. Ceramic samples were cut into bars. Some bars were powdered for microwave absorption measurements (the absorption measured in the bars before powdering was nearly the same). The samples studied by microwave absorption, ceramic pieces (bars), powders or the twinned crystal, were placed in a 2.5 mm diameter × 250 mm long EPR grade quartz tube. The twinned crystal was selected from the powders and glued to the end of a quartz rod. All samples were flushed and the EPR tube filled with helium gas at atmospheric pressure, and sealed to prevent sample deterioration from contact with oxygen and water vapour.

The superconductivity and superconducting transition temperature T_c of our samples were determined by standard dc four-probe measurements (silver paint and pressed indium contacts were utilized), Meissner effect measurements using a Hewlett-Packard model 428BR flux-gate magnetometer, and our microwave absorption method [5] using a Brüker ER 200-SRC EPR spectrometer operating at an X-band with a TE_{103} cavity. The microwave measurements were made in the same way as ordinary EPR except that in this case the measured absorption is not magnetic resonance. The EPR quartz tube containing the superconducting sample was placed in a position of a maximum microwave magnetic field (a minimum electric field) in the resonant cavity, which was critically coupled (minimum reflectivity) with the input/output waveguide. A dc magnetic field plus a small ac modulation field (at 100 kHz) were applied and the modulated part of the reflected power from the resonant cavity was detected by a phase sensitive amplification at the frequency of the modulation field. The demodulated signal was then digitized and recorded by a computer. The field modulation technique used gave rise to a derivative-like signal when the modulation amplitude exceeded approximately 1 G. Lower modulation amplitudes resulted in dramatically different results, depending on the direction of the dc magnetic field scan.

To facilitate the measurements near zero magnetic field, the magnet of the EPR spectrometer was charged by an external constant-current power supply connected to the rapid scan soils of the spectrometer. The temperature was regulated and controlled by an Air Products helium flow system and measured by means of Au(Fe)-chromel thermocouple.

III. RESULTS

Due to different conditions in their preparation (different conditions during the annealing in oxygen) we have found (see Fig. 1) that for the sample A (B) the onset of the superconducting transition was 101 K(97 K) and that a zero resistivity state was achieved at 93 K (74 K). The samples also differ markedly in the width ΔT of the superconducting transition.

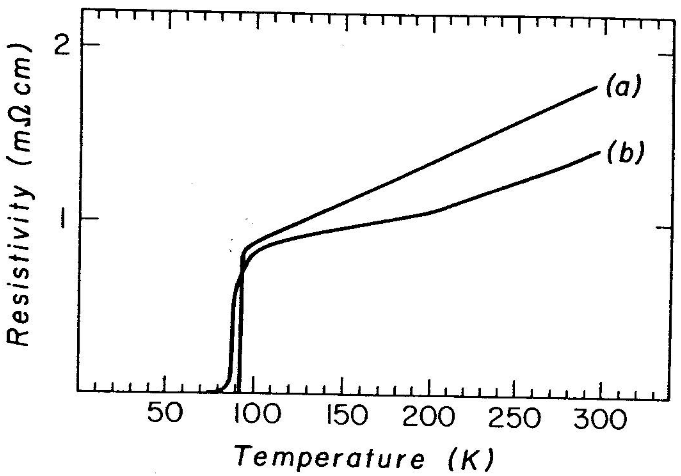

Fig. 1. The temperature dependence of the resistivity. (a) Sample A, (b) sample B.

As the samples are cooled through T_c a strong microwave absorption is observed for magnetic fields of up to at least 12 kG. Figure 2 shows the modulated (i) low-field absorption, which can be seen in detail in the inset for different modulation amplitudes, and (ii) broad absorption, which is present up to the highest field measured. The results that we present were obtained with 2.5 G modulation amplitude.

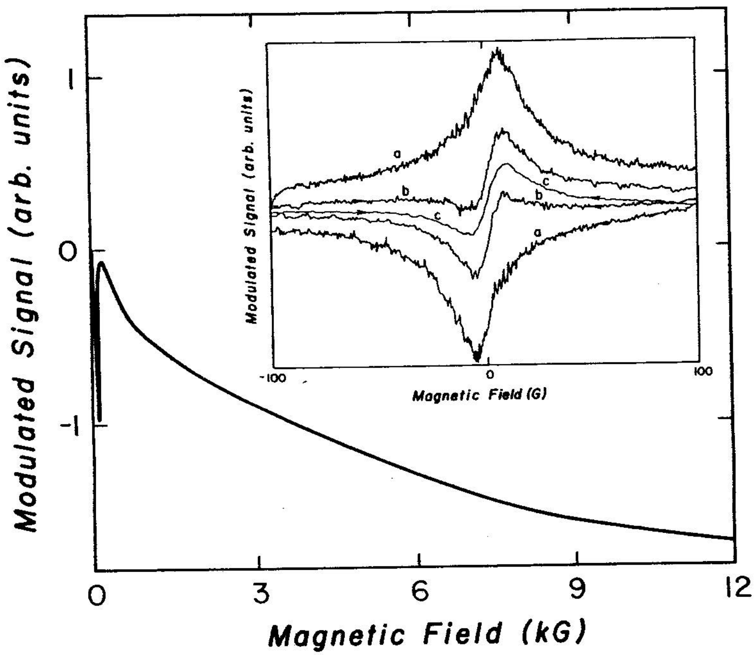

Fig. 2. The modulated microwave absorption in superconducting $YBa_2Cu_3O_7$ versus magnetic field at 30 K. The inset shows the modulated lowfield absorption (negative fields are applied in the opposite direction to positive fields): (a) modulation amplitude of 1.25×10^{-2} G, (b) modulation amplitude of 2.5×10^{-1} G. (c) modulation amplitude of 2.5 G. Arrows indicate the direction of the field scan.

The amplitude (peak-to peak intensity) of the low-field absorption is plotted as a function of temperature for samples A and B in Fig. 3. We have found [5] that the precipitous increase of the low-field signal amplitude as one cools below the superconducting transition provides an accurate measure of the T_c. It follows immediately from Figs. 1 and 3 that the narrower superconducting transition in sample A manifest itself by a sharper temperature dependence of both the resistivity and the low-field absorption. It should be noted that the sensitivity of microwave absorption measurements covers about five decades and is superior in that way, in detecting the onset of the superconductivity, to the resistivity as well as to the low frequency ac susceptibility measurements. Our measurements show that the low-field absorption is found in large ceramic pieces (e.g. bars), powdered ceramics, and in single grains ($\sim 10 \times 100 \times 100\ \mu m^3$) of $YBa_2Cu_3O_7$, which are probably twinned single crystals.

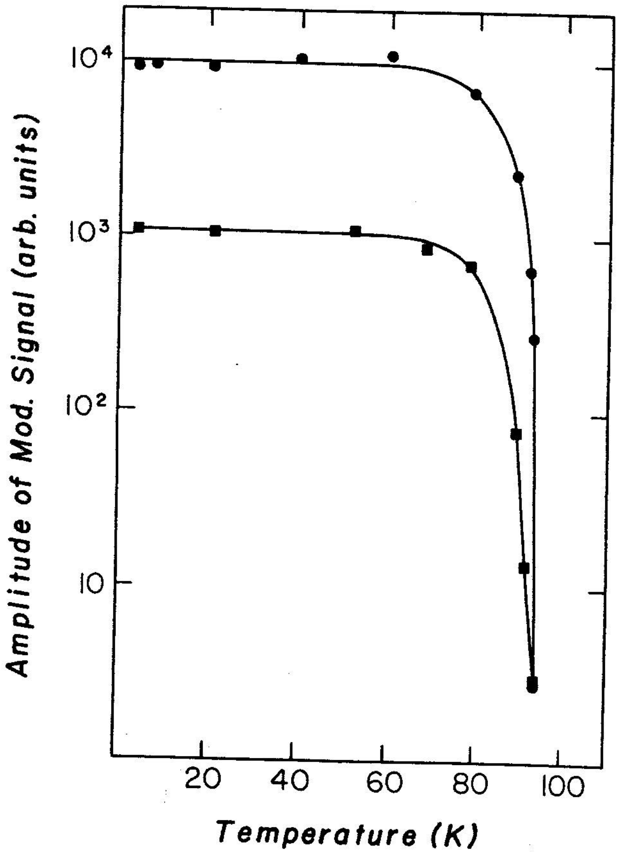

Fig. 3. The temperature dependence of the amplitude of the low-field absorption. ● = sample A, ■ = sample B.

The temperature dependence of the broad signal amplitude (as a measure of the amplitude we take the integral under the modulated signal versus magnetic field up to the maximum field measured 12 kG) has the same qualitative behaviour as the low-field absorption, i.e., a precipitous increase at temperatures

near T_c and then a flattening out at lower temperatures. It is very important to mention that we are measuring a modulated absorption signal not absorption itself and that there appear to be two distinct features in the non-resonant microwave absorption. Monitoring the Q of our cavity we have found, in agreement with others [15, 17, 18], a minimum in the microwave absorption near zero magnetic field. The higher the value of the scanning magnetic field, the larger the microwave absorption. In one of our earlier publications [19] it was shown that the field-cooling has a similar effect on the low-field absorption: the larger the magnetic field in which the sample is cooled, the larger the remanent absorption of microwaves at zero field and the broader the low-field absorption.

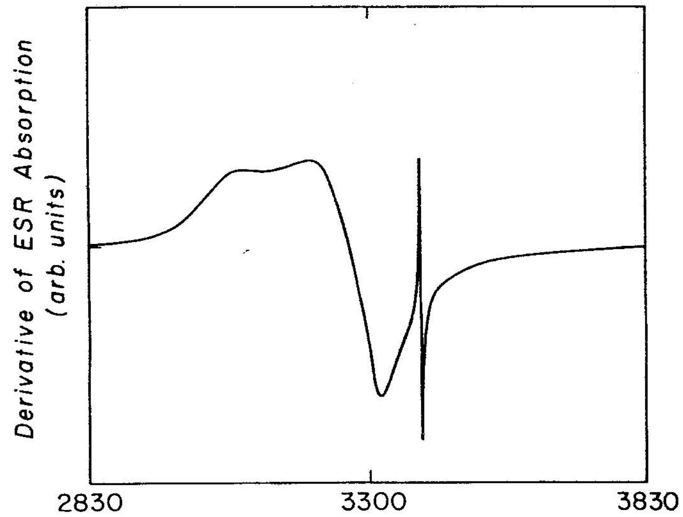

Fig. 4. Room temperature Cu^{2+} EPR line in $YBa_2Cu_3O_7$ altogether with an EPR standard (DPPH).

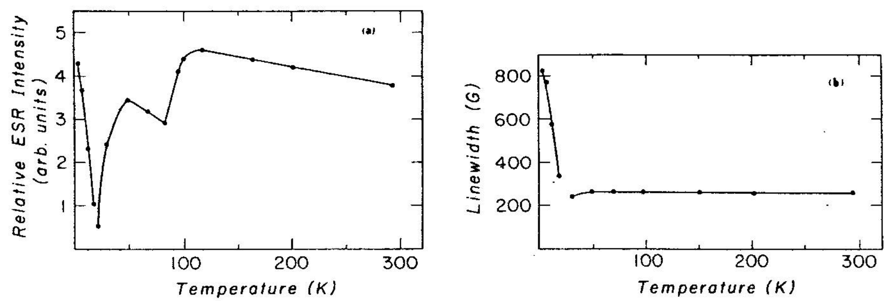

Fig. 5. The temperature dependence of the Cu^{2+} EPR signal intensity (a) and linewidth (b).

All investigated samples exhibit a weak paramagnetic resonance signal associated with Cu^{2+} ions in sites with axial symmetry ($g_{\parallel} = 2.206 + 0.004$, $g_{\perp} = 2.048 + 0.002$) of the neighbouring O^{2-} ions. In our samples at room temperatures the spin density is $\leqq 10^{18}$ cm^{-3}. Typical results of the EPR experiments are shown in Figs. 4 and 5.

We used an EPR cavity to expose the sample of $YBa_2Cu_3O_7$ to microwave radiation and to measure simultaneously the microwave induced dc voltage across a small bar of the sample, $10 \times 1 \times 1$ mm^3 (voltage contacts in a standard four probe technique were utilized) when the dc bias current I_{dc} is zero. Since thermal EMF's of several μV can easily develop in these materials, the samples were allowed to reach a steady state at all temperatures. We found a striking magnetic field dependence of the microwave induced dc voltage V_{dc}; the voltage peaks at zero magnetic field and then it decreases gradually to zero for both field sweep directions in a slightly asymmetric manner (Fig. 6). Should be noted that

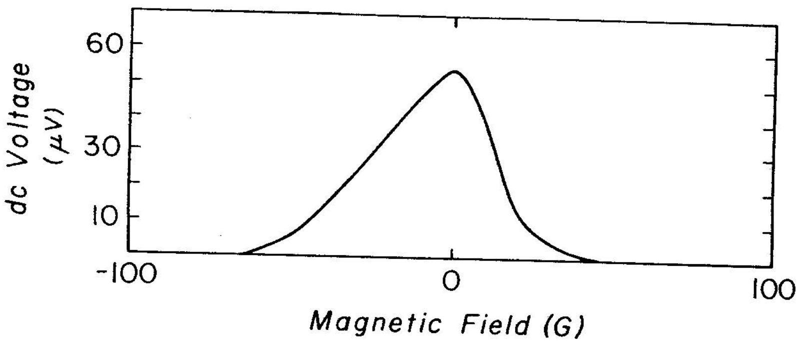

Fig. 6. Magnetic field dependence of the microwave induced dc voltage across the sample at 30 K, $f = 9.42$ GHz and $P = 63$ mW. Negative fields are applied in the opposite direction to positive fields.

in similar experiments on small and long Josephson junctions of regular low T_c materials [20, 21] with in-line current bias the polarity of the induced voltage did reverse when the magnetic field was reversed; however, dependent on the bias and on how it is injected into the junction, a complicated behaviour can be expected. The power and temperature dependencies of the microwave induced dc voltages are shown in Figs. 7a and 7b, respectively. It is clear from an inspection of Fig. 7 that the above mentioned dependencies are linear to a good approximation. The power dependence of the induced voltage tends to saturate at the highest microwave power that we used (approximately ~80 mW).

It should be noted that the induced dc voltage across the sample is detected only when the sample is superconducting; that correlation was checked by simultaneous resistivity and microwave absorption measurements.

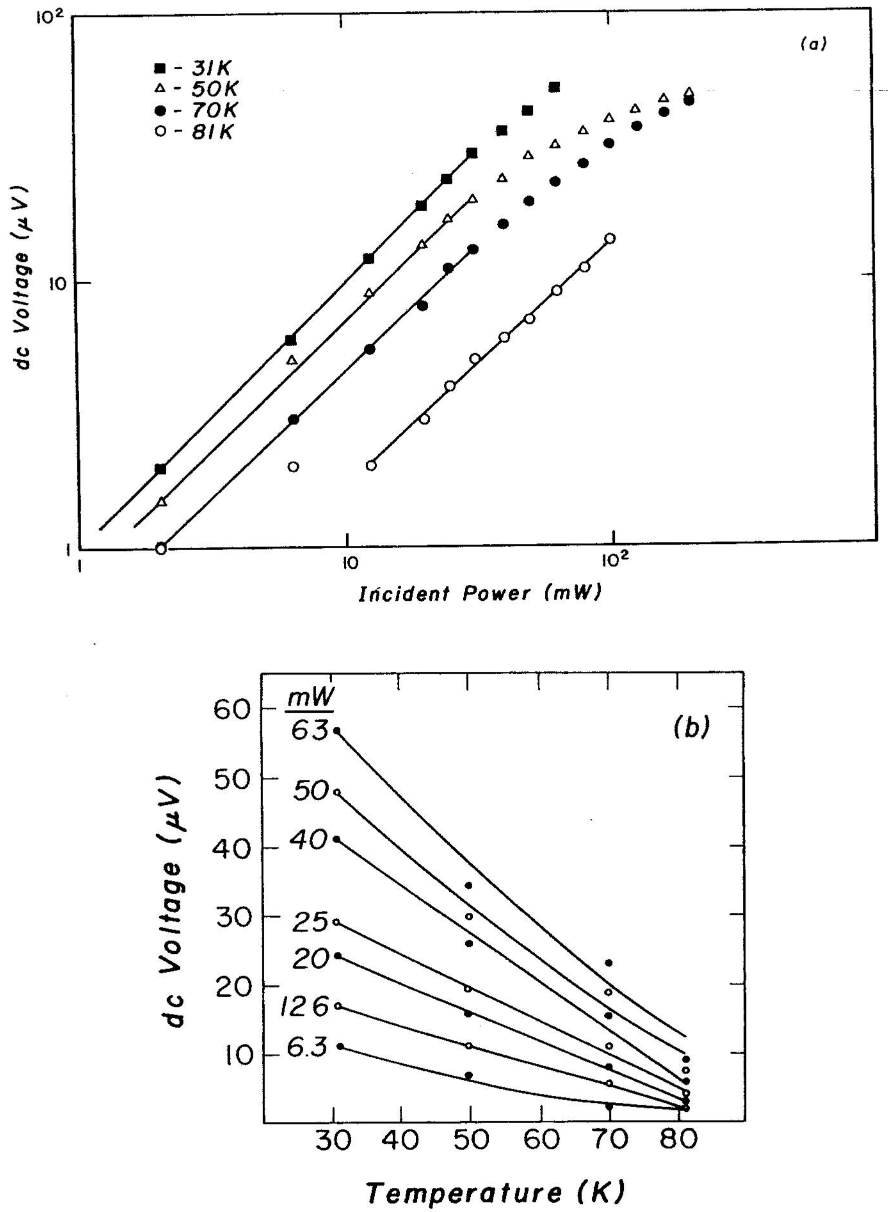

Fig. 7. (a), (b) The power and temperature dependence of the microwave induced dc voltage, respectively.

In order to demonstrate that the low-field absorption is present also in other high-T_c compounds, Figs. 8a, b show such signals for $La_{1.9}Sr_{0.1}CuO_4$ and $BaPb_{0.75}Bi_{0.25}O_3$, respectively.

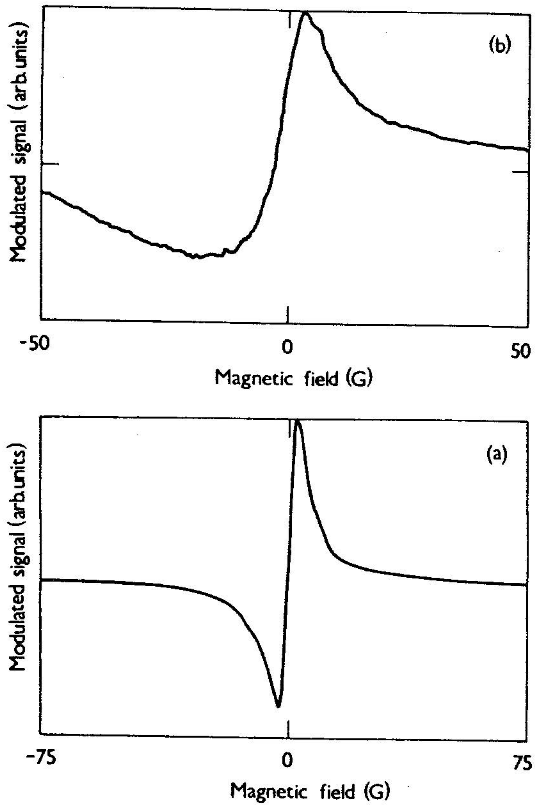

Fig. 8. The modulated microwave absorption at 4 K with 2.5 G modulation amplitude in superconducting (a) $La_{1.9}Sr_{0.1}CuO_4$ ($T_c = 25$ K) and (b) $BaPb_{0.75}Bi_{0.25}O_3$($T_c = 11$ K). Negative fields are applied in the opposite direction to positive fields.

IV. DISCUSSION

Both the EPR line intensity and the linewidth exhibit a very weak temperature dependence going from room temperature down to ~ 100 K. This behaviour corresponds rather to the Pauli-like than the Curie-like paramagnetism. Below T_c the EPR signal gradually diminishes and at low temperatures (~ 18 K) a new line appears with a Curie-like behaviour.

The measurements of EPR spectra give the most precise information on the electronic ground state — in the case of Cu^{2+} ions a distincition between a $d_{x^2-y^2}$ and a d_{z^2} ground state. The latter is indicated by a low g-value ($g < 2.04$), the former by an axial spectrum with $G = (g_{\parallel} - 2)/(g_{\perp} - 2)$ approximately equal to 4.0. Our results indicate the $d_{x^2-y^2}$ electronic ground state of the Cu^{2+} ions.

It should be mentioned that according to us the observed EPR signal in $YBa_2Cu_3O_7$ does not originate in the 90 K superconducting phase. This is in agreement with the conclusion presented in our first paper [5] and also in agreement with the EPR results of several other groups, e.g. the results of Albino et. al. [22] concerning the $YBa_2Cu_3O_7$ single crystals.

Since the low-field absorption is also observed in single grains of $YBa_2Cu_3O_7$ (we have been able to obtain resolved signals from as small an amount of the sample as $\sim 10^{-3}$ mg), we conclude that it is mainly an intragrain effect and not due exclusively, for instance, to intergrain current loops or intergrain Josephson junctions. In ceramics the intergrain effects can, of course, contribute to the absorption.

It was found that the orthorhombic phase is heavily twinned in contrast to the tetragonal phase in which such twins are absent and are impossible owing to crystal symmetry. Deutscher and Müller [23] have recently proposed that the twin boundaries introduce intragrain Josephson junctions in high-T_c oxides as a result of a very short superconducting coherence length in these materials. Intragrain current loops circulating among the array of these junctions formed at the many twinning (and grain) boundaries may then lead to a dissipative, glassy behaviour [23].

Although the exact structure of various twins is not known, according to Deutscher and Müller [23] they are essentially regions of the tetragonal phase about one unit cell thick, which have not been transformed into the orthorhombic phase. The oxygen content of the orthorhombic phase at room temperature is about 6.7—7 atoms per formula unit. The high temperature (nonsuperconducting) tetragonal phase, which can be sustained at low temperatures by quenching, has an oxygen content close to 6.5 per formula unit. There are strains which are necessary to match the two halves of the crystal, and at the twin boundary an inhomogeneity occurs which consists of a local redistribution of the oxygen and the oxygen-vacancies, and local angular distortions. It is conceivable that the twin boundaries form electronic potential barriers, which act as Josephson junctions, because of (a) the sensitivity of the structure to tetragonal-to-orthormobic distortion, (b) the low carrrier density (of the order of $\sim 10^{21}$ cm^{-3}), and (c) the tendency toward localization caused by inhomogeneities.

Although there has been much discussion about naturally occurring Josephson junctions and arrays in samples of high-T_c materials, not many data exists to support this view. Recently [24] direct measurements of the current carrying properties of single grain boundaries in $YBa_2Cu_3O_7$ have shown that the currents in the grain boundaries can be weaker by several orders of magnitude when compared to the currents in the grains. These experiments support our model for the behaviour of these materials in microwaves at low magnetic fields.

The behaviour of Josephson junctions in small magnetic fields was studied theoretically by de Gennes [25]. Khachaturyan et al. [14] proposed that the large change in diamagnetism over a narrow magnetic field range and the low-field absorption in high-T_c superconductors can be understood within that theory. On the basis of their microwave resistance and reactance measurements Portis et al. [15] concluded that the low-field microwave induced superconducting currents. Another explanation, which associates the low-field signal with changes in the diamagnetic susceptibility upon the transition from the Meissner state to the mixed state, has also been proposed [9].

So far we have mentioned the non-resonant microwave absorption data only on ceramic samples. The first microwave absorption measurements on single crystals of $YBa_2Cu_3O_7$ were reported by Bogachev et al. [26]. They have found that for a high modulation amplitude the intensity of the modulated signal has a maximum and the linewidth a minimum at B parallel to the crystal c-axis. At $B \perp c$ the relation between the intensity and the linewidth is reversed. For small both dc magnetic fields and modulation amplitudes the observed signals exhibit a strongly oscillating character. For repeated scans the oscillatory behaviour is preserved in the same region of magnetic fields around the zero field. The obtained results suggest the presence of processes of the Josephson nature.

Blazey et al. [27] have also observed the microwave absorption in single crystals of $YBa_2Cu_3O_7$ for a 10 G field sweep and found it to be composed of series of very narrow (~ 2 mG), uniformly spaced absorption lines that appear to be associated with the mixing of flux states. The modulated envelope is dependent upon the microwave power and similar to that observed in ceramic samples, suggesting that measurements on ceramics may represent in part an integrated powder average of such series. The line spectrum is indeed converted into the spectrum observed in ceramics by increasing the field modulation amplitude by two orders of magnitude. The single crystals of $YBa_2Cu_3O_7$ exhibit prominent striations, which may be associated with domain boundaries, primarily along a single [110] direction and separated by a distance of ~ 1 μm. According to Blazey et al. [27] fluxons within these boundaries are the source of the absorption lines. Microwave absorption also in single crystals arises from viscous fluxon damping [15] and is assumed to occur when the microwave power is sufficient to stabilize the mixing of the adjacent flux states.

Our model, providing the corresponding flux states, contributes towards the

understanding of the origin of the non-resonant microwave absorption in both ceramic and single crystal high T_c superconductors. It was first proposed in a preliminary communication [28] and distinguishes two regimes. At first, starting at $B = 0$, the absorption is minimal and then, as the magnetic field increases, the absorption increases rapidly. Samples can be considered as made of many long Josephson junctions (one dimension of such junctions $L > \lambda_J$, where λ_J is the Josephson penetration depth). This penetration depth [29, 30] is equal to $(\Phi_0/\mu_0 d J_c)^{1/2}$, where J_c is the critical current density of the junction and d its magnetic thickness ($2\lambda_L + t$, where t is the natural thickness of the barrier). Since λ_L is large in these materials (> 100 nm), d is essentially $2\lambda_L$. For barriers at grain boundaries, twins, etc., such current densities exist that barrier dimensions $L > \lambda_J$ occur. Estimates of the upper bound of λ_J for typical ceramic Y-Ba-Cu-O samples give values of $\lambda_J \sim 1$—5 μm, while grain sizes are of order 10—20 μm; thus in many grains the ccondition $L > \lambda_J$ can exist. An external field greated than

$$B^J_{c_1} = \frac{4}{\pi} \mu_0 J_c \lambda_J \tag{1}$$

will then create Josephson fluxons in the barriers [29, 30]; the fluxons can be nucleated by an external field or the field due to currents flowing through the junction. The microwave fields will then move the fluxons along the junctions. The motion of fluxons in a barrier is influenced by friction due to surface losses in the barrier and due to interactions with normal electrons. These losses provide a natural mechanism for dissipation; consequently fluxon motion in long junctions leads to a voltage across the barrier. As the external magnetic field increased, more fluxons are created leading to more losses. The critical fields $B^J_{c_1}$ from equation (1) ranges from around 0.1 G down to much lower values depending on demagnetizing factors. In the second regime of our model, above a certain magnetic field Abrikosov vortices can start, being nucleated in the bulk of the material. These vortices are also moved around by the microwaves. The cores of the Abrikosov vortices are normal (unlike the Josephson fluxons) and there is dissipation. This changeover is seen as a break in the field dependence and is typically ~ 10 G. Such a low $B^*_{c_1}$ for the onset of the Abrikosov vortices is not in contradiction with the quoted B_{c_1} as low as 50 G or less since demagnetizing factors have to be taken into account.

As for the broad component of the magnetic field dependent microwave absorption, which dominates above approximately 30 G, we suggest that this component arises from the surface impedance with a possible contribution of pinned flux tubes [31].

The origin of the microwave induced dc voltage is also in the Josephson junctions; we propose that it can be associated with the reverse ac Josephson effect. A voltage V across a junction causes the phase of the order parameter to vary in time according to the ac Josephson relation. As an external magnetic field is applied, the junctions lose coherence and eventually switch off in a few Gausses. Actually the I-B_{ext} curve of the junction current in an external magnetic field shows a similar dependence for long junctions, in contrast with the Frauenhoffer diffraction pattern for small junctions.

The microwave induced dc voltage was first observed for small junctions made of regular superconductors by Langenberg et al. [20]. Hamasaki et al. [21] found a peculiar magnetic field dependence of the induced voltage for long Josephson junctions. More recently Chen et al. [32] have observed during their microwave studies on Y-Ba-Cu-O high-T_c superconductors that the induced voltage is changing in a random fashion with both the microwave power and the frequency and that the polarity of the induced voltage is not only as a function of power, but that the polarities can be opposite also for two slightly different frequencies. They ascribed the induced dc voltage to the reverse ac Josephson effect.

It should be noted, however, that in view of the above mentioned experimental results we can not exclude the possibility that the microwave induced dc voltage across the superconducting $YBa_2Cu_3O_7$ ceramics is a result of some sort of rectification effects due to asymmetric I—V characteristics. On the other hand only very little is known about the hebaviour of a random network of long Josephson junctions in a static magnetic field and microwave radiation and the experimental results on long Josephson junctions of regular superconductors are hardly decisive.

V. SUMMARY

The magnetic field dependent absorption of microwaves provides us with an extremely sensitive and simple leadless method for the characterization of the high-T_c superconductors and the detection of possibly higher T_c phases in new samples. The microwave absorption and the microwave induced dc voltages measured across the sample can be explained on the basis of the existence of arrays of long Josephson junctions formed at detects, grain boundaries, and twins in the high-T_c superconductors. These Josephson junctions the fluxon motion in low magnetic fields, and in higher fields Abrikosov vortices can form in the bulk of the sample. Both types of fluxon motion lead to dissipation.

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ПОГЛОЩЕНИЕ МИКРОВОЛН И ПЕРЕХОД ДЖОЗЕФСОНА В СВЕРХПРОВОДЯЩЕМ

Приведены результаты измерения поглощения микроволн в диапазоне температур от 4 К по T_c в магнитных полях до 12 кГ. Наблюдено сильное поглощение в заввисимости от магнитного поля, что дает возможность создания подходящей (независимой от размеров образцов) высокочувствительной методики применимой при исследовании свойств новых высокотемпературных сверхпроводников и поиске фаз с высшей T_c. Показаны также характеристики поперечного постоянного напряжения индуцированного микроволновым излучением в зависимости от величины магнитного поля. Максимальное напряжение наблюдено при отсутствии магнитного поля. Предлагается модель поглощения слабых полей с зависимостю индуцированного напряжения на поглощении микроволн.