PHYSICAL REVIEW B VOLUME 42, NUMBER 4 1 AUGUST 1990

Weak-link structure in $YBa_2Cu_3O_7$ single crystals: A microwave study

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(Received 8 December 1989)

Field-dependent microwave absorption in high-quality $YBa_2Cu_3O_7$ single crystals shows that even very low magnetic fields penetrate into the sample. In the temperature interval of a few degrees below T_c, a broad hysteretic signal is observed. We interpret it in terms of a dense structure of weak links, whose origin is in twin boundaries and other thin defect planes. As the temperature is reduced, the critical currents of these junctions increase rapidly, and approach the bulk value. The broad signal then vanishes. At low temperatures one can observe narrow discrete absorption lines due to occasional defects of a different kind, which are thicker and, therefore, maintain a weak coupling. The dense structure of weak links, active just below T_c, may preclude the measurements of some intrinsic bulk superconducting properties close to T_c, and invalidate a straightforward comparison of experimental data with theoretical predictions.

I. INTRODUCTION

Measurements of the microwave properties of high-T_c oxide superconductors has fundamental physical and technological relevance. These measurements can, in principle, address the fundamental questions about the energy gap, its anisotropy, and the presence or absence of nodes in the energy gap $\Delta(k)$, as predicted by various proposed pairing mechanisms. To this end, it is essential to measure the microwave response of a homogeneous bulk superconductor. If the sample contains many Josephson junctions, or other weak links, they may dominate in the microwave response, so that the intrinsic superconductor properties cannot be determined. For example, the frequency dependence of the microwave absorption in sintered ceramic samples of $YBa_2Cu_3O_7$ was found to be like that of an *s*-wave superconductor.[1] However, this measurement could not be taken as conclusive, because it was also found that the magnitude of the absorption was orders of magnitude larger than the predictions of Bardeen-Cooper-Schrieffer (BCS) theory, and had to be attributed to the losses in intergranular Josephson junctions. A response closer to the intrinsic property of the superconductor, should be expected from single crystals and thin films. Indeed, the microwave absorption in single crystals[2] and thin films[3] was measured to be much smaller than in ceramic samples. Interestingly, the temperature dependence of the absorption deviated from the predictions of BCS theory. However, it was also observed that the level of absorption in single crystals of $YBa_2Cu_3O_7$ could vary considerably between the samples of different quality.[2] Various inhomogeneities in the samples could still produce a weak-link structure, and bring about an increased microwave absorption. The question is whether the absorption observed in the best available samples is still mainly due to remaining weak links, or one has reached the limit of a true bulk superconductor response. This can be conveniently tested if the microwave absorption is measured as a function of an external magnetic field. For a bulk type-II superconductor, one does not expect a significant change in the absorption until the lower critical field H_{c1} is reached. Above H_{c1}, a significant field penetration occurs, and the microwave absorption increases rapidly. If regions of weak superconductivity are present in the sample, the field will penetrate there even much below the bulk H_{c1}. The microwave absorption is expected then to be field dependent even at very low magnetic fields. Early measurements on $YBa_2Cu_3O_7$ single crystals[4–7] revealed a pronounced variation of the microwave absorption starting practically from zero field. The intensity and the form of the signals were strongly temperature dependent. Just below the superconducting transition temperature T_c, one could observe a broad signal whose intensity peaked at a few degrees below T_c, and then decreased rapidly as the temperature was further reduced.[4,5] At low temperatures one could detect a series of discrete lines.[7,8] Both types of signals were attributed[5,8] to Josephson junctions which may occur in single crystals.[9] The question remained: how could the signals with such different shapes and temperature dependences originate in the Josephson junctions? The main objective of the present paper is to elucidate this problem. We present microwave signals observed in single crystals of higher quality than those studied before.[5,10] The analysis of the results leads us to propose a model of a temperature-dependent weak-link structure. It occurs even in the best available single crystals, and this fact may have far-reaching consequences in various other measurements near T_c.

II. EXPERIMENT

Magnetic-field-dependent microwave absorption was detected by means of a Bruker ER 200D EPR spectrometer operating at 9.3 GHz (X band). The sample was placed in the center of a TE_{102} resonant cavity where the microwave magnetic component is maximum. The microwave field induces currents on the surface of the sample, and we monitor the resulting absorption. The sample could be rotated around a vertical axis, which is also the axis of polarization of the microwave field. The static field was in the horizontal plane. Two schemes of detection could be used. In the first, we applied a field modulation and lock-in detection of the modulated microwave absorption. Alternatively, we detected the absorption as a function of the field without a modulation. In this case we used repetitive field scans and accumulation of absorption signals by an external data system. The temperature was controlled by a helium-flow cryostat (ESR-10 Oxford Instruments).

The single crystals of $YBa_2Cu_3O_7$ were grown from copper-rich eutectic melts[11] in zirconia crucibles. These crystals are much less contaminated than those grown in alumina crucibles, which were studied before.[5,10] The zero-field microwave measurements of crystals grown in ZrO_2 crucibles showed that these crystals were of a very good quality,[2] so it was intriguing to examine whether weak links were still present.

Figure 1 shows a series of signals obtained with field modulation and lock-in detection. The microwave field was in the *ab* plane, while the dc magnetic field was along the *c* axis. In the absence of hysteresis, the signal trace represents the first derivative of the microwave absorption with respect to the applied dc field. Figure 2 shows a

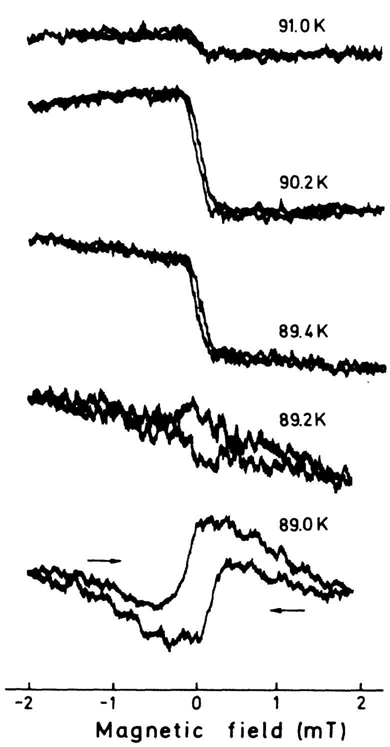

FIG. 1. Modulated microwave absorption in $YBa_2Cu_3O_7$ single crystals for $H \| c$. Each signal is recorded in forward and reverse field scans.

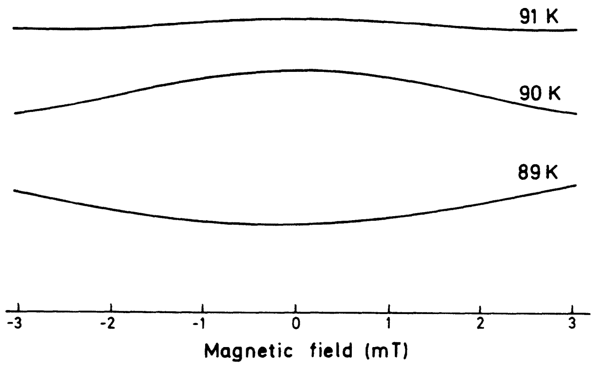

FIG. 2. Microwave absorption in $YBa_2Cu_3O_7$ single crystals for $H \| c$. The curves show transition from zero-field maximum- to zero-field minimum-type signals.

few traces of the absorption curve taken without field modulation and lock-in detection. As the temperature is reduced, one observes first a signal with a maximum at zero field. This signal increases over the temperature interval of about one degree and then decreases rapidly, being replaced by a signal with a minimum at zero field. The former signal is quite unexpected in superconducting systems. One might speculate that this signal could be a manifestation of superconducting fluctuations above T_c. However, when the crystal was freshly annealed in air at 450 °C, this signal was not observed. We have to conclude that its nature is not clear at present, and we shall not consider it in the next section where we propose a weak-link model for the behavior below T_c.

The superconducting signal with a minimum at zero field shows a hysteresis when the modulation scheme is used (cf. Fig. 1), but not in the directly detected absorption curve (cf. Fig. 2). This kind of hysteresis is due to the boundary current induced in the sample when the field is swept.[12] Figure 3 shows the evolution of the modulated absorption signal when the temperature is reduced. After an increase, the amplitude of the signal is gradually reduced. It is interesting to observe the signal traces in the forward and reverse field scans. The arrows in Fig. 3 indicate the positions at which the field sweep is reversed. At first, the signal recorded in the forward direction is retraced, and then there is an exponential-like approach to the signal level of the extended reverse field sweep. At lower temperatures, the initial reversible section increases, and the hysteresis vanishes together with the disappearance of the signal. No signal was observed at still lower temperatures.

When the crystal is rotated by 90°, the dc magnetic field penetrates into the *ab* plane. Figure 4 shows a series of signals recorded at various temperatures. The peculiar signal with a maximum at zero field, was not observed in this orientation. In fact, as the crystal is rotated from $H \| c$ to $H \| ab$, this signal decreases in amplitude. At $H \| ab$, only the superconducting signal with a minimum absorption at zero field is detected. Its amplitude also peaks at a few degrees below T_c as in the case of $H \| c$, but the hysteresis is almost absent.

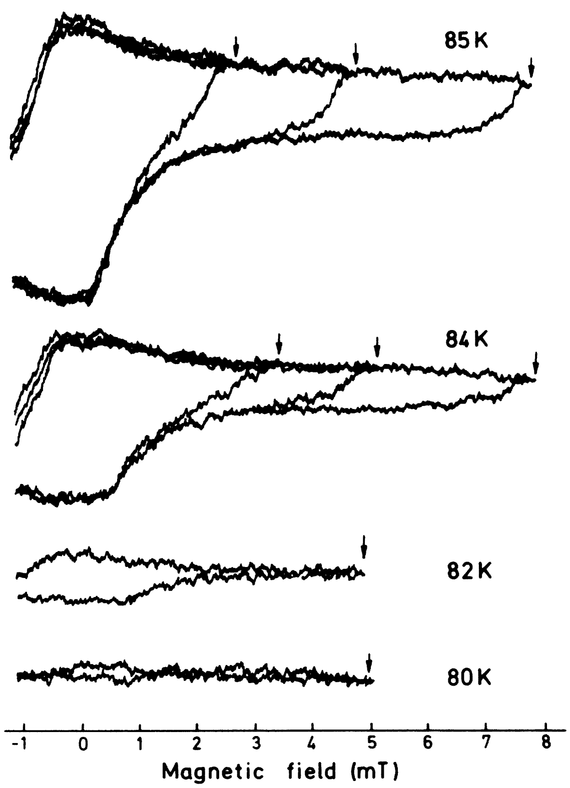

FIG. 3. Evolution of modulated microwave absorption signals at temperatures below those in Fig. 1. The arrows indicate the positions at which the field sweep was reversed.

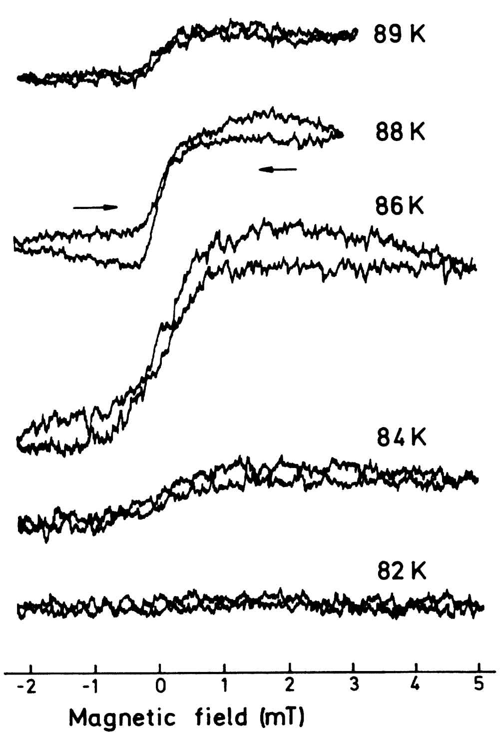

FIG. 4. Modulated microwave absorption signals in $YBa_2Cu_3O_7$ single crystals for $H \| ab$.

In the preceding experiments, the microwave field was always in the *ab* plane. If the crystal is mounted so that the microwave field is perpendicular to the *ab* plane, one observes the signals as in Fig. 4, but with an order of magnitude larger amplitude. This can be understood since the crystals have a platelet form, so that the cross section for the microwave flux is small when the microwave field is in the *ab* plane, and large when the microwave field is perpendicular to the *ab* plane. When the temperature is further reduced, one can observe discrete narrow lines similar to those reported earlier.[7,8,13,14] An important feature of these lines is the existence of a microwave power threshold above which they appear. In the present crystals this threshold is rather high. No lines could be detected when the incident microwave power was below 200*e* μW. Figure 5 shows signals observed at several high microwave power levels. The ap-

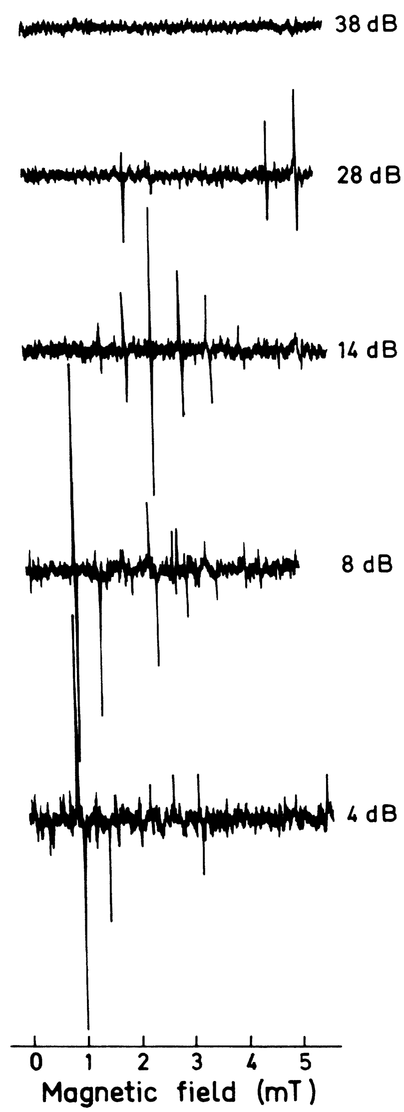

FIG. 5. Discrete microwave absorption lines (first derivatives) in $YBa_2Cu_3O_7$ single crystals for $H \| ab$ at 2.6 K. The attenuation of the microwave power below 200 mW is indicated.

pearance of new lines is observed as the microwave power is increased. Once present, the lines broaden at still higher power levels in the same fashion as reported previously,[14] and then disappears below the noise. The discrete lines observed in the present crystal do not form long periodic sequences, which is another point of distinction from the observations in previous crystals.

III. MODEL

The observed field dependence of the microwave absorption in $YBa_2Cu_3O_7$ single crystals indicates that even very low fields can penetrate into the sample. This feature cannot be explained on the basis of Abrikosov vortices in a conventional type-II superconductor. Rather, it suggests that a weak-link structure is present in the sample. The usual idea of a single crystal as a perfect uniform body excludes any possibility of weak links. However, real crystals can have defect planes, inhomogeneities due to a contamination during crystal growth, twin boundaries, microscopic cracks on the edges, and other kinds of imperfections. The question is whether such thin nonsuperconducting regions could noticeably affect the superconducting state.

Deutscher and Muller[9] have analyzed the case of a superconductor-insulator-superconductor (SIS) junction. Just below T_c, the critical current of the junction is proportional to the product of the gap values Δ_{SI} at the two SI interfaces, i.e.,

$$I_c(T)=\frac{\pi}{2eR_n k_B T}\Delta_{SI}^2(T) , \tag{1}$$

where R_n is the normal resistance of the junction. The relation of Δ_{SI} to the bulk value Δ is determined by the coherence length ξ, and the lattice parameter a. One can distinguish two limiting cases:

$$\text{(i)}\quad \frac{\xi^2(0)}{a\xi(T)} \ll 1, \quad \Delta_{SI}(T)\approx\frac{\xi^2(0)}{a\xi(T)}\Delta(T)\ll\Delta(T) ,$$

$$\text{(ii)}\quad \frac{\xi^2(0)}{a\xi(T)} \gg 1, \quad \Delta_{SI}(T)\approx\Delta(T) .$$

Case (i) is always realized close to T_c since $\xi(T)$ diverges. The critical current of the junction is then much smaller than the bulk critical current, i.e., the insulating barrier perturbs the superconducting state giving rise to a weak link in the system. One may expect that samples of classical superconductors may also have imperfections in the form of thin nonsuperconducting barriers. However, a weak-link behavior near T_c has not been observed. The reason can be found if one considers the temperature interval of case (i)

$$\frac{T_c-T}{T_c}\le\left[\frac{a}{\xi(0)}\right]^2 , \tag{2}$$

which, for classical superconductors with $a/\xi(0)\approx10^{-3}$, and $T_c\approx10$ K, yields $(T_c-T)\approx10^{-5}$ K, i.e., an interval within the transition width. Hence, the gap value at the interface rises to the bulk value practically at T_c. If the barrier is very thin (small R_n), its critical current will be close to the bulk value, so that no weak-link feature could be observed. However, one may expect a different behavior in high-T_c superconductors because of their short coherence lengths. In particular, for $YBa_2Cu_3O_7$ one has $\xi_{ab}(0)=34$ Å, and $\xi_c(0)=7$ Å.[15] Thus, for a barrier perpendicular to the *ab* plane, the temperature interval in Eq. (2) can be estimated with $a/\xi_a(0)\approx10^{-1}$, and $T_c\approx10^2$ K to be $(T_c-T)\lesssim1$ K. For a barrier parallel to the *ab* plane (tunneling along the *c* axis), one has $c/\xi_c(0)\approx1$ and the temperature interval of Eq. (2) can be much larger. Therefore, we may expect to observe a weak-link phenomenology in high-T_c single crystals, in particular close to T_c.

In order to explain the present microwave signals, we propose the following weak-link structure of $YBa_2Cu_3O_7$ single crystals. In a temperature interval of a few degrees below T_c, there is a multitude of active weak links in the sample. All of the twin boundaries and other defect planes take part in this structure. The critical current of the junctions increases quadratically with temperature $I_c\propto(T_c-T)^2$ due to both, decrease of $\xi(T)$, and increase of the bulk gap $\Delta(T)$. Since these junctions are very thin, their critical currents approach the bulk value, and the weak-link manifestation vanishes. In addition, the crystal may have a small number of defects of a different kind, which have a larger R_n, and preserve their weak-link character even at lower temperatures. In that range, the critical current still rises, but at a much slower rate according to the relation[15]

$$I_c(T)=\frac{\pi\Delta(T)}{2eR_n}\tanh\left[\frac{\Delta(T)}{2k_BT}\right] . \tag{3}$$

The preceding proposed weak-link structure in single crystals can explain the observed microwave signals. Just below T_c, the multitude of active weak links causes the crystal to act like a granular superconductor. The similarity of the signals in Figs. 3 and 4 to those in sintered ceramic samples[16] should be noted. The latter, however, do not vanish at lower temperatures. This is not surprising because the microwave absorption in ceramic samples is dominated by intergranular junctions which are much thicker than the defect planes in single crystals, and retain their weak-link character even at very low temperatures. The microwave absorption in a granular superconductor can be treated by considering the microwave response of a single representative junction.[17] If the sample is exposed to a dc field H_0, and a microwave field $H_1\cos\omega t$, appropriate currents are induced on its surface. For a junction on the surface, driven by a dc current I_0 and a microwave current $I_1\cos\omega t$, the response is determined by the equation

$$\frac{C\hbar}{2e}\frac{d^2\phi}{dt^2}+\frac{\hbar}{2eR_n}\frac{d\phi}{dt}+I_c\sin\phi=I_0+I_1\cos\omega t , \tag{4}$$

where C is the capacitance, R_n the normal resistance, and I_c the critical current of the junction.[18] In the absence of the microwave field, the phase would adjust to an equilibrium value ϕ_0 given by $I_c\sin\phi_0=I_0$. In a complicated network of interconnected junctions, the phases will tend to arrange so that the coupling energies are minimized. Thus, each junction may have a different I_0. As the dc field is swept, there will be rearrangements of phases, i.e., phase jumps at individual junctions.[19] These processes will give rise to an increased noise in the microwave signals. However, the broad feature of the signals can be explained if an averaged current $I_0 < I_c$ is assumed. The nonlinear equation (4) cannot be solved analytically. However, for small microwave currents only small oscillations will be induced

$$\phi(t)=\phi_0+\phi_1(t), \quad |\phi_1| \ll |\phi_0| \tag{5}$$

so that one can obtain a linear equation in $\phi_1(t)$, whose solution is straightforward. The capacitive term can usually be neglected in very small junctions, and the absorbed power becomes

$$P=P_n\frac{1}{1+\eta} , \tag{6a}$$

$$P_n=\tfrac{1}{2}I_1^2R_n , \tag{6b}$$

$$\eta=\frac{I_c^2\cos^2\phi_0}{\left[\dfrac{\hbar\omega}{2eR_n}\right]^2} . \tag{6c}$$

The critical current of a Josephson junction is reduced when a magnetic field is applied, and this will have an effect on η and P. The critical current can have nodes at certain values of the field. However, we have to consider absorption in a large ensemble of junctions in the sample which, upon averaging, yields a monotonic decrease of the critical current with field. Thus, Eqs. (6) explain the observed absorption minimum at zero magnetic field. At high fields, the absorption reaches the value P_n, which is simply the loss in a normal junction.

The technique of field modulation and lock-in detection does not yield a simple derivative of Eq. (6a). If a small modulation field $H_M\cos\omega_M t$ is superimposed on H_0, one can replace I_0 in Eq. (4) by $I_0+I_M\cos\omega_M t$, and the solution for modulated absorption signal, lock-in detected at ω_M, becomes[17]

$$S_M \sim I_c(1+\eta)^{-3/2}\left[-\frac{dI_c}{dH}H_M+I_M\sin\phi_0\right] . \tag{7}$$

The first term is the field derivative of the absorption curve. It does not depend on the direction of the field sweep. The second term changes sign when the sweep direction is reversed because the induced boundary current changes the sense of flow. This term gives a hysteresis in the observed signals. In ceramic samples, the change occurs in less than 0.01 mT following the point of sweep reversal.[12] Figure 3 shows that in single crystals for $H_0\|c$ the transition takes place within an interval of 1 mT or larger. The initial retracing of the forward scan shown in Fig. 3 indicates that the reversal of I_0 is not a simple linear function. The hysteresis observed for $H\|ab$ (cf. Fig. 4) is much smaller. Within the present model, it could be explained as a consequence of an easier field penetration, or equivalently, a smaller I_0 which flows partially along the c axis. Therefore, ϕ_0 is smaller in this case, and the contribution of the second term in Eq. (7) is reduced.

Finally, one has to consider the temperature dependence of the signals. When the temperature is lowered from T_c, the critical current increases rapidly, and so does the signal given by Eq. (7). In that range, the critical current is still small, and the fields penetrate through the whole junction length, i.e., the junction can be classified as a small junction.[18] However, as the critical current of the junction approaches the bulk value, the field penetration becomes restricted to the edge region and is finally eliminated. The derivative term in Eq. (7) then vanishes. Also, when I_c becomes much larger than I_0, the phase ϕ_0 decreases to zero, and the second term in Eq. (7) vanishes, too.

When the temperature is further reduced, only a small number of weak links remain. It is no longer appropriate to assume an averaged boundary current I_0. In the limit of a single junction in a superconducting loop, one can expect a periodic opening of the junction as the field is swept, which gives rise to a discrete set of absorption lines. This can also happen if one of the junctions is much weaker than the others. If, however, several junctions have comparable strengths, the situation can be more complex. The mechanisms of the microwave absorption have been treated by other authors,[13,20,21] and shall not be considered here. We note only that, in the single crystals studied here, the microwave-power threshold was considerably higher than in the previous studies.[8,13,14] It might be that the critical currents of the remaining weak links were relatively higher in the present crystals.

IV. DISCUSSION

From the analysis of the field-dependent microwave absorption, we have concluded that even the highest quality single crystals of $YBa_2Cu_3O_7$ have a weak-link structure. Within a few degrees below T_c, the structure is very dense, and originates probably at twin boundaries and other thin defect planes. At lower temperatures these links become strong and practically indistinguishable from the bulk material. An occasional small number of defects of some other kind gives rise to links that remain weak even at low temperatures.

The implications of the weak-link structure in single crystals can be considerable. Thus, in the measurements of the zero-field microwave surface resistance as a function of temperature, one has to take into account that within a few degrees below T_c the microwave absorption will be dominated by the weak links. Therefore, any comparison with the predictions of BCS, or another theory for bulk superconductors, is not appropriate for that temperature region. Only at temperatures below this region one can safely compare the experimental data with theoretical predictions. The remaining weak links are active absorbers of microwaves only at particular values of the applied external field. It is very unlikely that the sample will be cooled just in the right field for the activation of one of the weak links. Even if this happens, one can avoid the effect of the weak link if low microwave power levels are used so that the threshold for the link activation is not reached.

The weak-link structure can modify other superconducting properties of single crystals. Hylton and Beasley[22] have shown recently that a sample with a weak-link structure has an effective-field penetration depth λ_{eff} which can be different from the London penetration depth λ_L for the bulk material. The difference is small when the superconducting domains are large compared to the London penetration depth, but may become significant for small domains. The latter case is realized in $YBa_2Cu_3O_7$ single crystals within a few degrees below T_c. In a flux penetration experiment one measures the effective penetration depth. Since it depends on the critical current of the weak links, the temperature dependence of the measured penetration depth may reflect the temperature dependence of the junction critical current more than that of the intrinsic penetration depth of the material. At lower temperatures, where only a small number of weak links may remain, the size of the domains is larger so that the measured penetration depth is practically equal to the intrinsic London penetration depth.

In conclusion, single crystals of $YBa_2Cu_3O_7$ have a dense weak-link structure in the temperature interval of a few degrees below T_c. This structure may profoundly influence some measured superconducting parameters, such as the microwave surface resistance, the field penetration depth, and possibly transport properties and critical fields. Therefore, the experimental data from this temperature range should not be straightforwardly compared with the predictions of various theories of bulk homogeneous superconductors. It seems that only at lower temperatures one can confidently compare theory with experiment.

ACKNOWLEDGMENTS

This work was supported by the Cornell Materials Science Center and by National Science Foundation Grant No. CHE-87-03014.

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