# Size of Josephson junctions in Ba-Y-Cu-O compounds

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Microwave-absorption measurements using a conventional electron-spin resonance spectrometer provide a novel leadless technique for the detection of Josephson junctions and their size in superconductive Ba-Y-Cu-O compounds.

Soon after the exciting discovery of superconductivity above 90 K in a mixed phase Ba-Y-Cu-O compound,[1] it became known that the superconductive phase was oxygen-deficient perovskite $Ba_2YCu_3O_{9-y}$.[2] It had zero resistance at 91 K with a transition width of only 1.4 K. Currently, large efforts are being made in many laboratories in order to find empirically whether still another phase with an even higher $T_c$ can be prepared. Some recent reports are promising in this respect.[3,4] In parallel, investigation of all relevant structural, electromagnetic, and spectroscopic parameters is carried out with the hope that their knowledge will help to elucidate the mechanism of superconductivity and guide the future quest for new materials.

In view of the granular structure of these sintered compounds it was suggested that the observation of zero resistance was due to a continuous path of weakly coupled grains between the leads used in the experiment. With this hypothesis, Josephson effects could be expected, and, indeed, they were recently reported.[3,5] In this paper we report on a novel technique for the detection of Josephson effects in which no external leads on the sample are required. It is based on a very sensitive microwave absorption measurement using a conventional electron-spin resonance (ESR) spectrometer. With a sweep of the external dc magnetic field, one observes the well-known diffraction effect of the Josephson junctions.[6] Thus, we were able to get the first direct experimental evidence of the size of these junctions in Ba-Y-Cu-O compounds. Also, we have observed an interesting hysteretic behavior.

Our measurements were carried out on the samples of both multiphased "green" $Ba_{0.8}Y_{1.2}CuO_4$, and monophase "black" $Ba_2YCu_3O_{9-y}$. With a variation in the sintering conditions, we have prepared samples with the same nominal composition, but different temperature dependencies of the resistivity.

The samples were prepared starting from the mixture of appropriate amounts of oxides $Y_2O_3$, CuO, and $BaCO_3$, followed by firing in air at 900 °C, grinding and forming into pellets. The black superconducting samples of $Ba_2YCu_3O_{9-y}$ ($T_c$=91 K) were obtained when the sintering temperature was 980 °C. An additional treatment in the stream of oxygen was also used to improve the superconducting transition (width 1.5 K).[7] When sintering was performed at a lower temperature (700 °C), the samples with the same nominal composition exhibited semiconductor-type behavior of the resistance at lower temperatures. Green superconducting multiphase samples of $Ba_{0.8}Y_{1.2}CuO_4$ were obtained with the sintering temperature of 860 °C. It was observed that a very small change of this temperature, or of the sintering time, could result in nonsuperconductive samples. In particular, the sample sintered at 900 °C had resistivity characteristics of an insulator.

Conventional ESR spectrometer (Varian E-109, 9 GHz) was used for the microwave absorption measurements. The only modification was the introduction of an externally fed coil for a constant magnetic-field offset so that sweep across zero field could be performed. Due to a high $Q$ value of the resonant cavity, and elaborate phase-sensitive detection, ESR spectrometers are suitable for the detection of a very small change in the microwave absorption.

At temperatures above $T_c$, no signal could be observed, even with the highest gain on the ESR spectrometer. As the temperature was lowered to $T_c$ and below, one could observe a large signal as shown in Fig. 1. It has the form of the first derivative of the actual variation of the microwave absorption due to the use of the audio-frequency modulation and first harmonic lock-in detection which is standard in ESR spectrometers. The observed signal does not originate from the usual spin resonance. It is centered at zero magnetic field, and, moreover, has a negative sign. The latter has been established by comparison with the

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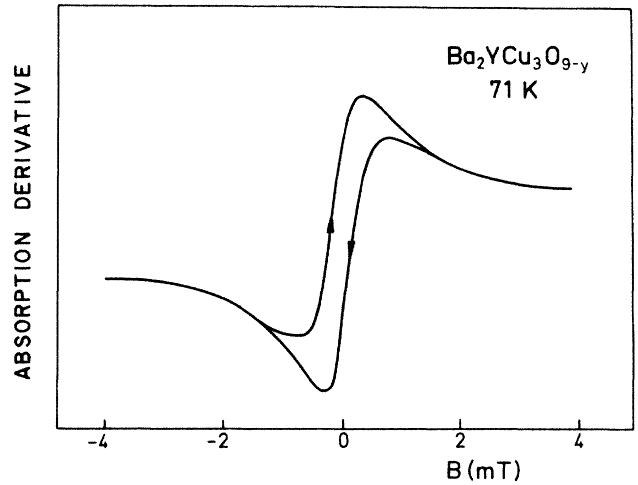


FIG. 1. Absorption line (first derivative) of a superconductive sample of $Ba_2YCu_3O_{9-y}$ at 71 K.

signal of the reference sample diphenylpicrylhydrazyl (DPPH). Thus, the variation of the microwave absorption shows a minimum at zero magnetic field, and increases to a constant value as the magnetic field is swept away from zero. The hysteretic behavior will be discussed below.

The depth of the absorption variation, proportional to the peak-to-peak intensity of the derivative signal, has an interesting temperature dependence. Figure 2 shows the curves of the signal intensity and resistance for two different black $Ba_2YCu_3O_{9-y}$ samples. In the superconductive sample, the drop in the resistance correlates perfectly with the rise of the signal intensity. The latter increases by several orders of magnitude, and remains almost constant at lower temperatures. The signal could also be observed in the other black sample with a semiconductor resistance behavior, but is much weaker. Weak diamagnetism could be detected in susceptibility measurements on this sample.[8] One may assume that the superconductive phase is present in a small concentration so that its grains may form clusters, but do not form an uninterrupted path between the leads used in resistivity measurements.

Figure 3 shows the signal intensity and resistance as a function of temperature for the superconductive green multiphase sample $Ba_{0.8}Y_{1.2}CuO_4$. The superconducting transition is broader (about 10 K) in this sample. The initial signal rise correlates well with the resistance drop. However, as the temperature is lowered further, the signal intensity continues to increase, though at a lower rate, and finally saturates. This behavior is probably due to the multiphase character of the sample. For obvious reasons, no further information could be obtained from resistivity measurements once the zero resistance is reached. The second green multiphase sample with an insulator property, showed no signal at all.

The observed absorption signal can be explained in the following way. The oscillating microwave field (magnetic component) impinges on the sample and induces surface

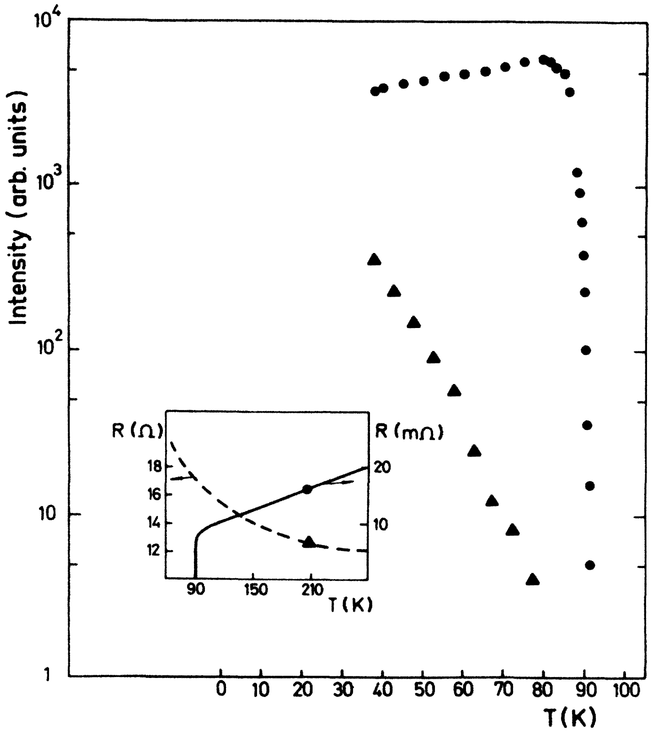


FIG. 2. Temperature dependence of the absorption line intensity of two differently sintered samples of $Ba_2YCu_3O_{9-y}$. The inset shows their resistance characteristics.

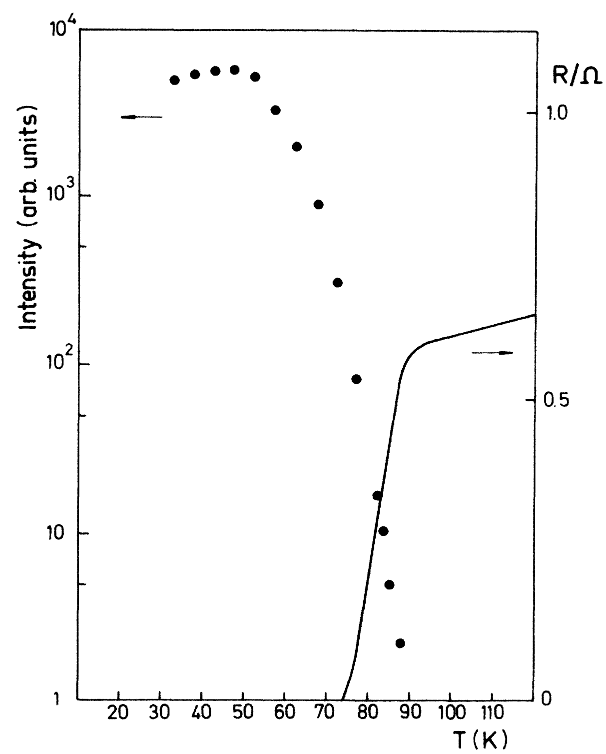


FIG. 3. Temperature dependence of the absorption line intensity and resistance of a superconductive multiphase sample $Ba_{0.8}Y_{1.2}CuO_4$.

currents, which, in their turn, give rise to reflection and partial absorption of the microwaves. In the normal state of the sample, the absorption level does not change when the sample is exposed to a relatively weak dc magnetic field. The situation, however, changes when the sample becomes superconductive. At zero external dc magnetic field, the supercurrents can flow from grain to grain via Josephson junctions, and provide screening for the penetration and absorption of the microwaves. When the dc magnetic field is increased from the zero value, the maximum supercurrent through a Josephson junction is reduced according to the formula[9]

$$I = I_0 \frac{\sin(\pi\phi_j/\phi_0)}{\pi\phi_j/\phi_0} , \qquad (1)$$

where $I_0$ is the maximum current in the absence of the magnetic field, $\phi_j$ is the magnetic flux through the junction, and $\phi_0$ is the flux quantum. As a consequence, the supercurrent screening effect is reduced, and one observes an increased absorption of the microwaves. The diffraction formula (1) predicts also attenuated oscillations at higher magnetic fields. These are not observed in our experiment due to a distribution in the size of the Josephson junctions on the surface of the sample. An average junction size can be estimated from the value of the dc magnetic field at which the absorption line is at half height. Taking the penetration depth of the magnetic field to be 500 Å on each side of the junction, one obtains the value of 20 $\mu$m for the average length (diameter) of the junctions.

We have observed some difference in the diffraction pattern between various samples. In general, good superconducting samples have a narrower diffraction pattern. Since the intensity of the observed line is also larger in these samples, one may conclude that the superconducting phase is not only present in a higher concentration, but also is formed in larger grains. In order to check the validity of the conclusions, we have ground one of the superconducting samples into powder, and then measured it with the present method. The diffraction pattern was, indeed, much broader, indicating that the remaining Josephson junctions were much smaller in size.

The hysteretic behavior shown in Fig. 1 is another interesting effect manifested in the present method. Note the asymmetry in each of the sweeps. Approaching the zero field does not yield the same shape as after crossing it and leaving towards a higher field. The sample keeps some memory of the preceding field. In the superconducting black samples, the hysteresis gradually diminished at lower temperatures. We suggest that in these samples the hysteretic behavior could be due to the recently proposed vortices in a system of Josephson coupled grains.[10] In the multiphase green samples, the hysteresis behavior is more complicated, which may indicate that the nonsuperconductive phases also play a role in the magnetism of the sample.

In conclusion, we have shown that the study of the microwave absorption as a function of an external dc magnetic field can be used as a new leadless technique for the detection of Josephson junctions in granular superconductors. Their size could be determined from the width of the diffraction pattern. In our samples of Ba-Y-Cu-O compounds, the average size of the Josephson junctions was found to be 20 $\mu$m in diameter. Good superconducting samples exhibited somewhat narrower diffraction pattern, and a larger signal intensity, which reveals that the superconducting phase was not only present in a greater concentration, but also formed larger grains. The present method detects the onset of superconductivity in excellent correlation with the resistivity measurement. Moreover, it reveals further evolution at lower temperatures where the resistivity measurements give no additional information. This feature could be particularly useful in the study of multiphased samples. Due to a high sensitivity of the present method, it could also become useful for the detection of a small concentration of a possibly higher-temperature superconductive phase in new samples.

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