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Thermal detection of microwave absorption in high-temperature superconductors

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Nonstationary processes of microwave absorption in a superconducting ceramic sample of Y-Ba-Cu-O have been studied. A method of direct measurement of microwave absorption by detection of the sample-temperature changes versus an external magnetic field has been designed. A thermodynamical description of the studied phenomena is proposed as a generalization of the model of local temperature T^* of the Josephson-junction system in granular superconductors. The results are in good agreement with the predictions of the model of weakly linked superconducting clusters presented by Ebner and Stroud.

INTRODUCTION

Magnetically modulated microwave absorption (MMMA) observed in superconductors in low-intensity magnetic fields has been recently studied in a number of research centers[1–3] and applied to investigations of the properties of superconducting materials. In particular, the microwave-absorption dependence on magnetic-field intensity and the rate of its change, as well as the microwave power and temperature of the studied sample, have been the subject of thorough studies both in classical[4] and high-temperature superconductors. Most frequently, microwave absorption was measured on a standard electron paramagnetic resonance (EPR) spectrometer by applying the second modulation of magnetic field which enabled us to record the microwave absorption signal derivative with respect to magnetic field. Measurements of microwave absorption versus microwave power proved that the system of internal Josephson junctions formed between the grains of a ceramic sample is heated by the microwaves.[5] This fact permitted the introduction of the concept of a local temperature T^* of the system of internal Josephson junctions, which for high microwave powers is higher than the temperature T of the bulk sample. The interaction of microwaves with the internal system of Josephson junctions, as well as with a macroscopic sample, has been usually studied by an EPR spectrometer with the second modulation of magnetic field. A strong dependence of the microwave-absorption signal on the amplitude of the second modulation prompted many authors[6–11] to abandon the idea of using the second modulation in microwave-absorption studies. Pakulis and Osada [6] proposed a method of direct current measurements on a microwave detector, which permitted the determination of the microwave power reflected from the resonator containing the studied superconducting sample versus an external magnetic field and other parameters. This paper proposes a method based on direct thermal detection of microwave absorption (TDMA) which enables MA (microwave-absorption) measurements without the necessity of using a second modulation and, moreover, permits a better insight into the interaction of microwaves with the system of Josephson junctions (JJs) and the thermodynamic properties of granular samples of a high-temperature superconductor in a microwave field. Previously the technique of the thermal detection had been used only for electron paramagnetic resonance measurements,[12] but not for investigation of microwave absorption of superconductors. The results obtained by the method of TDMA can provide information on the sample properties of interest from the point of view of electronic applications.

EXPERIMENTAL DETAILS

A single-phase high-quality sample of superconducting $YBa_2Cu_3O_{6.9}$ was studied on a standard EPR RADIOPAN SE/X-2943 spectrometer at a frequency of 9.4 GHz. The system was cooled by liquid-nitrogen vapors flowing through a quartz Dewar. The ceramic sample in the shape of a sphere, 3 mm in diameter, was placed in a microwave resonator type TE_{102} at the maximum of the microwave field magnetic component. The magnet of the EPR spectrometer was replaced by Helmholtz coils driven by a generator and amplifier with a sawlike voltage, which permitted the measurements to be run in low magnetic fields at any sweep rate without any hysteresis (Fig. 1). The residual magnetic field was compensated by two additional Helmholtz coils so that the zero-field cooling (ZFC) could be performed in a magnetic field lower than 0.1 G. The above precaution was necessary as the position of the signal on the scale of the field B_0 and the width of MA signal depended on the magnetic field in which the superconducting sample was cooled below the critical temperature. MMMA was studied by applying the modulation of the magnetic field at a frequency of 100 kHz with an amplitude of a few gauss.[13] The sample was first cooled to the superconducting state in a field lower than 0.1 G and then an external magnetic field, modulation field, and microwave field were applied. The temperature of the sample was measured by a copper-constantan thermocouple mounted in a hole in the sample. This design of the measuring setup enabled simultaneous (on two recorders) registration of the MMMA signal and the sample temperature (TDMA signal) as a function of an external slowly varying magnetic

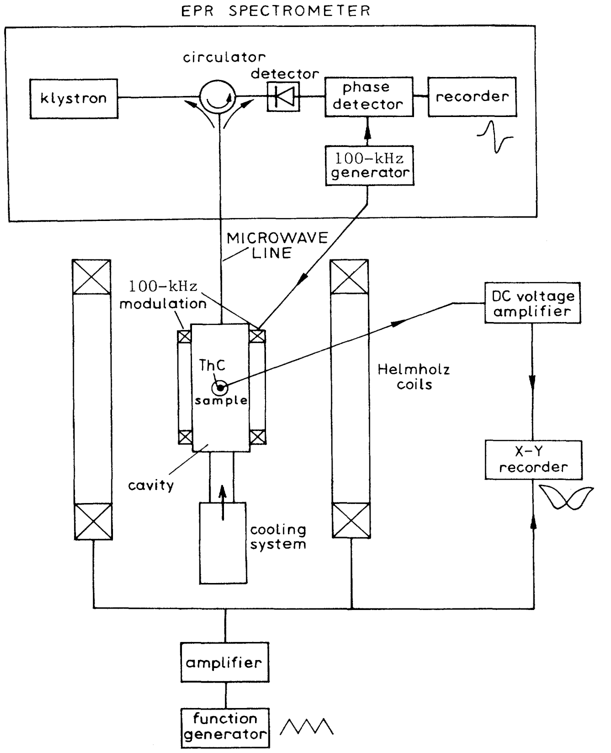

FIG. 1. Experimental setup for simultaneous measurements of the MMMA and TDMA.

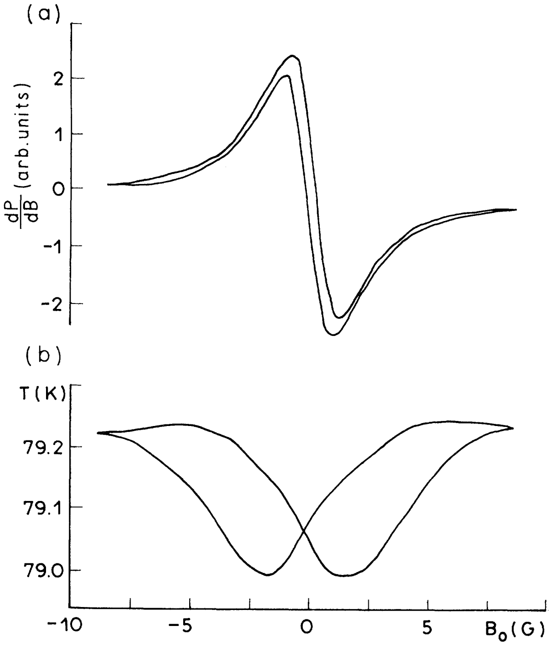

FIG. 2. (a) A signal of magnetically modulated microwave absorption (MMMA), (b) thermal detection of microwave absorption (TDMA).

field. The recording of TDMA signal required the thermocouple dc voltage amplification by about 120 dB on a highly stable KIPP amplifier, which was then supplied to the Y input of an X-Y type recorder. It should be mentioned that for thermal detection of microwave absorption by using a thermocouple, the thermocouple itself, as well as the sample and the bath, are also heated to a small degree. This effect is responsible for the increase of the background of the recorded signal; it is not sensitive to small changes in the magnetic field. A good indication of the microwave phenomenon in the sample is its relative temperature change versus the external magnetic field.

RESULTS

The design of the measuring setup enabled simultaneous recording during the experiment of the sample temperature changes [Fig. 2(b)] and the MMMA signal. The latter was typical of granular superconductors.[14,15] When the second modulation of magnetic field is applied, the MMMA signal is the first derivative of MA signal with respect to the magnetic field. The actual shape of MA signals of superconductors is characterized in contradistinction to a standard EPR signal, by a nonzero value in the whole range of magnetic field (until the critical intensity is reached) and shows a sharp minimum only in the vicinity of the zero magnetic field. The shape of the minimum resembles a reversed EPR line whose derivative is recorded on a standard setup for MMMA measurements [Fig. 2(a)]. This kind of dependence was recorded by Pakulis and Osada[6] who directly measured the current flowing through a microwave diode and could in this way determine the microwave power reflected by the resonator containing the superconducting sample studied versus an external magnetic field. However, since the microwave power reflected by the resonator is not related directly to absorption or emission of energy but only to a change in the Q parameter of the resonator depending on the coupling conditions, the problem of the sign of the recorded signal remains unsolved. The method of TDMA permits the solution of this problem by providing conclusive evidence that the minimum absorption occurs at $B_0=0$. Measurements of the temperature of the superconducting sample in microwave field revealed a strong dependence of the sample temperature on the intensity of an external magnetic field in the sample.

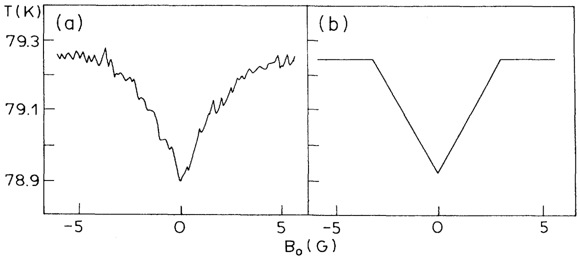

FIG. 3. (a) The sample temperature changes for quasistatic variation of magnetic field corresponding to the changes in JJs temperature, (b) the approximation of the real characteristic from (a) to simplify the computer modeling.

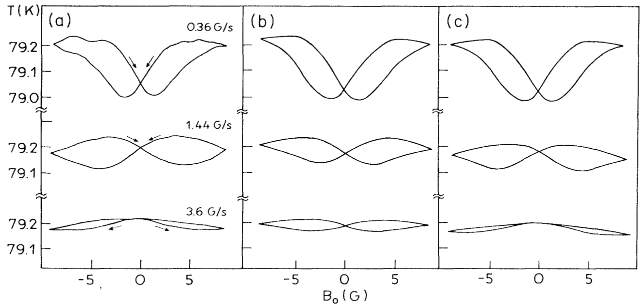

FIG. 4. (a) TDMA for three different rates of the magnetic-field changes, (b) the solution of Eq. (2) for the three different rates of magnetic-field changes, (c) the same solution with the phase shift induced by a finite value of thermal conduction between the sample and the bath.

At zero magnetic field the sample temperature reaches a minimum while for fields higher than 10 G, it becomes saturated and has a constant value of a few tenths of K higher than the minimum [Fig. 2(b)]. The course of the temperature changes, as the field is swept in both directions, and shows large hysteresis which depends on the rate of magnetic-field changes. The recorded TDMA signal was fully reproducible and, what is more important, it did not depend on the amplitude of the second modulation of the magnetic field. A signal of the same shape was recorded with the second modulation turned off. The dependence of the thermal detection signal on the rate of magnetic field changes was also analyzed [Figs. 3(a) and 4(a)]. For high rates of field changes the signal was smooth, the temperature changes were smaller, and the minimum of the signal was shifted towards higher values of magnetic field, which enhanced the hysteresis between the two directions of the field sweep. For quasistatic field changes, we could observe thermal fluctuations not detected at a high sweep rate. TDMA signal was recorded in different ranges of the external magnetic field sweep [Fig. 5(a)]. Upon decreasing the magnetic-field sweep range, the TDMA signal amplitude was observed to diminish. However, the character of the recorded signal remain unchanged. The thermal effects detected via a thermocouple did not occur above the critical temperature.

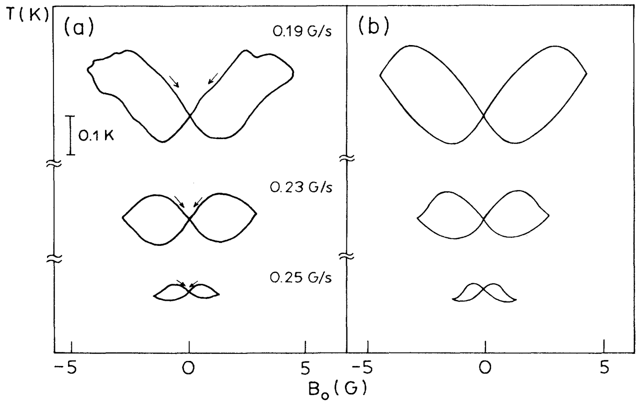

FIG. 5. (a) TDMA for different ranges of the magnetic-field sweep, (b) computer simulation performed according to Eq. (2).

THEORETICAL MODEL

The temperature changes of the studied granular superconducting sample subjected simultaneously to a microwave field of power P_0 and a magnetic field B_0 are related to the microwave absorption which occurs[1–3] after the sample has been cooled below the critical temperature T_c. Absorption which takes place in granular superconductors at microwave frequencies is, among other factors, due to the surface microwave impedance of the material, excitation of Cooper pairs to the state of quasiparticles, and in high magnetic fields also to viscous motion of fluxons. However, in low magnetic fields the dominant role is played by the losses taking place in the Josephson junctions formed at the contacts between the superconducting grains, and, in the case of monocrystalline samples, on the twin boundaries. In a microwave field of frequency ω, such a junction is polarized by the voltage $\tilde{V}=\omega\hbar/2e$, which causes the appearance of loss proportional to $\tilde{V}^2/R_n$, where R_n is the junction resistance in the usual model of a resistively shunted Josephson junction.[16] There appears also the emission of electromagnetic radiation of frequency $\omega_J=2\tilde{V}e/\hbar$.

Tilley[17] has developed a theory for arrays of interacting weak links. In this theory it is assumed that self-synchronization of an array can occur through the interacting of the links, which form the array with the common radiation field in the cavity. The effect of super-radiation appears as a consequence of interaction between the system of interacting junctions and electromagnetic field. In the "super-radiant" state, the array behaves essentially as a single weak link. Experimental results presented by Clark[8] show that such a point-contact array can enter the "super-radiant" state. Observations of coherent microwave radiation emitted by coupled Josephson junctions were reported by Finnegan and Wahlsten.[19]

Microwave power absorbed by a junction is mostly transformed into Joule heating which increases the temperature of the junction.[20] Superconductors are very poor thermal conductors well below T_C; the ratio k_n/k_s is about 10^2, where k_n and k_s are thermal conduction coefficients for the normal and superconducting state, respectively. Thus, a temperature gradient and the heat transfer it imposes appear between the superconducting grains and the system of junctions heated by microwaves. By analogy to the thermal detection of EPR,[12] an effective temperature T^* characterizing the system of junctions is introduced. The correctness of the model description involving the concept of the effective temperature has been supported by the observations reported by Stankowski, Czyżak, and Martinek.[5] In that paper, it was assumed that Josephson junctions formed among the grains of the superconducting sample in consequence of having absorbed microwave power reach a temperature

T^* different from that of the bulk sample T_S. In order to explain the dynamics of MA observed by TDMA, we shall make a generalization of the previously proposed model. Let us introduce four reservoirs (Fig. 6), which at a dynamic equilibrium for high microwave powers have $T^* \neq T_S$ and $T_S \neq T_B$, where T_B is a temperature of the bath. Thus we shall consider the following four reservoirs: (1) a microwave field of power P_0, (2) a system of Josephson junctions (JJs) of a temperature T^*, (3) a superconducting sample whose temperature T_S depends on the power P_0 and field B_0, and (4) a thermostat of great thermal capacity at a constant temperature T_B, which contains the studied sample.

The four systems (reservoirs) exchange the three fluxes of energy (a) from microwaves to JJs proportional to P_0, (b) from JJs to the bulk sample proportional to $T^* - T_S$, and (c) between the sample and the thermostat proportional to $T_S - T_B$.

The exchange of energy between the microwaves and JJs (flux a) occurs relatively fast (10^{-10} s).[21] The power absorbed by the JJs is part of the total microwave power $P = aP_0$. The exchange of energy between the JJs and bulk sample (flux b) is determined by the thermal conductivity, characterized by the thermal transfer coefficient k_1. The heat transport from the sample to the bath is determined by the thermal transfer coefficient k_2.

Dynamics of the heat flux flowing through the sample is described by the heat flow equations:

$$\operatorname{div}[k \operatorname{grad} T(\mathbf{r},t)] + g(\mathbf{r},t) = cd\frac{\partial T(\mathbf{r},t)}{\partial t} , \tag{1}$$

where k is the thermal transfer coefficient, $g(\mathbf{r},t)$ is the volume density of the heating power generated in the system, d is the density, and c is the specific heat of the material. For the system considered, we can assume as the boundary conditions that T_B=const. and that the heating power is generated in a region of a small size (Josephson-junction system) relative to the volume of the sample. Solving Eq. (3) within the approximation of the model of lumped parameters we get an expression describing the thermal response of the system (time dependence) $T_S(t)$ to excitation by the power of the following time dependence $P(t)$:

$$T_S(t) = T_B + \int_{-\infty}^{t} P(x)\frac{\partial Z(t-x)}{\partial t}dx , \tag{2}$$

where the function

$$Z(t) = (1/k_2 a_2)[1-\exp(-t/\tau_2)] \tag{3}$$

is the transient thermal impedance of the studied system and $\tau_2 = M/k_2 a_2$, where M is the thermal capacity of the sample and a_2 is a geometrical factor describing geometrical conditions of the heat exchange between the sample and the bath.

For slow changes of magnetic field, in the vicinity of the zero value of this field intensity, the sample temperature reproduces the resonancelike line of JJs. To simplify the computer modeling procedure, we have approximated this real function by the triangle

$$P(B_0) = \begin{cases} \alpha B_{\text{width}} & \text{for } |B_0| \geq B_{\text{width}} \\ +\alpha B_0 & \text{for } 0 \leq B_0 < B_{\text{width}} \\ -\alpha B_0 & \text{for } -B_{\text{width}} < B_0 < 0 , \end{cases} \tag{4}$$

where B_{width} is the field in which the sample temperature begins to be constant, and α is a coefficient which is a linear function of the power P_0. Both the experimental and the approximate characteristics are presented in Fig. 3.

The above characteristics are similar to the dependence of the internal energy of the JJs in a superconducting sample on the intensity of a constant magnetic field, following from the calculations by Ebner and Stroud [Figs. 1(c) and 4 in Ref. 22] for a model of the weakly linked superconducting clusters. In that model, superconducting grains make weak contacts and form closed loops which pass the superconducting current. Such a model is a good approximation of the structure of granular ceramic superconductors. The energy of the system comprising a large number of superconducting loops of different areas, in temperatures higher than zero, reaches a sharp minimum for zero magnetic field, while for the fields higher than $\approx 0.5(\phi_0)/b^2$ it reaches a constant value (parameter b is the grain diameter and ϕ_0 is a flux quantum).

Solving Eq. (2) for changes in the absorbed microwave power, depending on an external magnetic field as in Fig. 3(b), and having introduced $t = B_0/\dot{B}_0$, where $\dot{B}_0$ is the rate of the magnetic-field sweep, we obtained the diagrams shown in Fig. 4(b). A comparison between the experimental and theoretical data gave the relaxation time

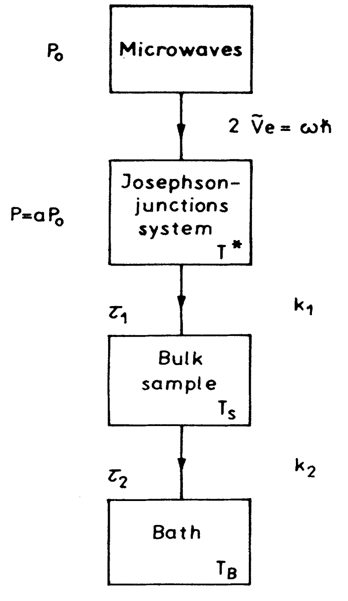

FIG. 6. Energy flow diagram. The four thermodynamical reservoirs interacting through microwave absorption by Josephson-junction and heat transport.

value of $\tau_2 \simeq 1$ s. For high rates of magnetic-field changes, the dependences presented in Fig. 4(b) differ from experimental ones by a shift in phase. To account for this we have introduced an additional factor δ into the exponent of Eq. (3) $(-t-\delta/\tau_2)$, which describes the shift in phase between the field and the sample response, and is related to the relaxation time τ_1 describing the processes of energy transfer between JJs and the sample (Fig. 6). Assuming $\delta = 0.95$ s we have obtained the diagrams presented in Figs. 4(c) and 5(b) which are in good agreement with the experimentally recorded dependencies. The shift in phase revealed only for high sweep rates is a consequence of a finite rate of energy exchange among the four thermodynamical systems considered. Our description has been found to be correct for all ranges of the magnetic-field sweep considered, which is confirmed in Fig. 5. A comparison between the dependencies obtained by the method of TDMA and those calculated by Ebner and Stroud[22] enabled us to estimate the mean area of clusters formed in the studied granular Y-Ba-Cu-O sample to be about 3 μm.[2]

CONCLUSIONS

The results of the studies of microwave absorption in polycrystalline $YBa_2Cu_3O_{6.9}$ carried out by the method of direct measurements of the sample temperature have shown that the following: (1) Microwave absorption is minimum at $B_0 = 0$ and then increases with the magnetic-field intensity up to the value of B_{width} reaching a constant value for higher fields; (2) The observed phenomena can be interpreted in terms of the model of weakly linked superconducting clusters; (3) The phenomena of the energy flow among the thermodynamical reservoirs characterized by the temperature T^*, T_S, and T_B provide a good explanation of the hysteresis in microwave absorption and thus of the related nonstationary processes of microwave absorption in granular superconductors.

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[1] R. Durny, I. Hautala, S. Ducharme, B. Lee, O. G. Symko, P. C. Taylor, D. J. Zheng, and I. A. Xu, Phys. Rev. B **36**, 2361 (1987).

[2] J. Stankowski, P. H. Kahol, N. S. Dalal, and J. S. Moodera, Phys. Rev. B **36**, 7126 (1987).

[3] K. W. Blazey, K. A. Mueller, I. G. Bednorz, W. Berlinger, G. Amoretti, E. Buluggiu, A. Vera, and F. C. Matacotta, Phys. Rev. B **36**, 7241 (1987).

[4] M. Warden, L. Baselgia, D. Berlowitz, P. Erhart, B. Senning, M. Stallder, G. Stefanicki, A. M. Portis, and F. Waldner, Proc. 24th AMPERE CONGRESS 1988, p. 727.

[5] J. Stankowski, B. Czyżak, and J. Martinek, Phys. Rev. B **42**, 10 245 (1990).

[6] E. J. Pakulis and T. Osada, Phys. Rev. B **37**, 5940 (1988).

[7] Y. Maniwa, A. Grupp, F. Hentsch, and M. Mehring, Physica C **156**, 755 (1988).

[8] M. Giura, R. Fastampa, R. Marcou, and E. Silva, Physica B **169**, 623 (1991).

[9] A. Gould, M. Huang, S. Tyagi, and S. M. Bhagat, J. Appl. Phys. **69**, 4880 (1991).

[10] F. J. Owens, Phys. Rev. B **43**, 8631 (1991).

[11] F. J. Owens, Physica C **178**, 456 (1991).

[12] J. Schmidt and J. Solomon, J. Appl. Phys. **37**, 3719 (1966); L. Janssens and J. Witters, *ibid.* **41**, 2064 (1970).

[13] B. Czyżak and J. Stankowski, Phys. Status Solidi B **160**, 219 (1991).

[14] A. Dulcić, and A. I. Veinger, Phys. Rev. B **42**, 2155 (1990).

[15] A. S. Kheifets and A. I. Veinger, Physica C **165**, 491 (1990).

[16] A. Barone and G. Paterno, *Physics and Application of the Josephson Effect* (Wiley, New York, 1982).

[17] D. R. Tilley, Phys. Lett. **33A**, 205 (1970).

[18] T. D. Clark, Phys. Rev. B **8**, 137 (1973).

[19] T. F. Finnegan and S. Wahlsten, Appl. Phys. Lett. **21**, 541 (1972).

[20] W. J. Skocpol, M. R. Beasley, and M. Tinkham, J. Appl. Phys. **45**, 4054 (1974).

[21] A. H. Silver and J. E. Zimmerman, Phys. Rev. **157**, 317 (1967).

[22] C. Ebner and D. Stroud, Phys. Rev. B **31**, 165 (1985).