RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 39, NUMBER 13 1 MAY 1989

Nonlinear microwave absorption in ceramic superconducting Y-Ba-Cu-O

Sergio M. Rezende and Flavio M. de Aguiar
Departamento de Física, Universidade Federal de Pernambuco, 50739, Recife, Brazil

(Received 11 July 1988; revised manuscript received 10 November 1988)

Novel microwave measurements in ceramic $YBa_2Cu_3O_{7-\delta}$ reveal that in the superconducting phase the absorption is highly nonlinear in the microwave power. At $T=80$ K the surface resistance increases with dc field at a rate of 0.0025 Ω/Oe, and with the microwave rf field at 0.07 Ω/Oe, resulting in an absorption that can be substantial in the absence of an applied dc field.

INTRODUCTION

The electrodynamic response of the ceramic high-T_c superconductor oxides has attracted increasing experimental and theoretical attention.[1-17] One of its most interesting aspects is the strongly field-dependent nonresonant microwave absorption observed at small values of the applied magnetic field (few Oe) and low microwave power (tens of mW).[7-14] This microwave absorption has been associated with thermally excited quasiparticles,[1,2,12] with damped fluxon motion,[9-11] or with Josephson junction currents.[8,14-16] Another interesting aspect of the electrodynamic response of the high-T_c materials is the harmonic generation observed with low-power radio-frequency driving[15] and with high-power microwave frequencies.[16,17]

In this paper we show that the microwave absorption of ceramic $YBa_2Cu_3O_{7-\delta}$ in the superconducting state is highly nonlinear in the microwave power. With a suitable balancing arrangement we demonstrate that the dependence of the microwave absorption on the microwave magnetic field is much stronger than on the biasing dc field. As a result the microwave loss at moderate power levels can be substantial even in the absence of a dc field.

EXPERIMENTS

The experiments were carried out at X-band microwave frequencies with the sample located at the bottom wall of a TE_{102} resonant cavity where the rf magnetic field is maximum. A dc biasing field H_0 was applied by a pair of Helmholtz coils either parallel or perpendicular to the microwave magnetic field $H_1 \sin\omega t$ and no effort was made to shield the Earth's field. The power was provided by a 2.2-W traveling-wave tube amplifier fed by a backward wave oscillator. Its frequency was stabilized by an external crystal oscillator mixer and manually adjusted to the resonance (9.4 GHz) of the slightly undercoupled cavity with $Q=2000$. A (0–50)-dB precision attenuator is used to vary the microwave power that is transmitted to the cavity by a ferrite circulator. The changes in the microwave response of the sample with varying H_0 or H_1 were measured with a balancing arrangement employing a hybrid-tee bridge illustrated in Fig. 1. The signal reflected from the cavity-sample system feeds one of the two symmetric arms of the tee while the other is fed by a reference signal derived from the incident radiation by a 10-dB directional coupler. In order to measure the nonlinear response, the amplitude and phase of the reference signal are adjusted at the minimum power level (20 μW) to produce a null in the detector arm and then the power is scanned. Since the reference signal is proportional to the incident power, the radiation that appears in the detector arm with increasing power arises solely from the changes in cavity reflection which are nonlinear in the microwave power. Alternatively we can balance the bridge at a given power level and scan H_0 to obtain the field dependence of the sample response, similar to the one measured with conventional ESR spectrometers.[8-13] The microwave output from the bridge was measured with a power meter (to give a direct measurement of the square of the cavity reflection coefficient) or detected with a local-oscillator heterodyne receiver for greater sensitivity.

All the results described in this paper were obtained with a sample of ceramic $YBa_2Cu_3O_{7-\delta}$, with resistive transition at $T_c=92$ K, in the shape of a slab with dimensions $1\times5\times10$ mm^3. Similar results were obtained with other samples of high-T_c ceramic materials. Figure 2 shows the variation of the microwave bridge output when the incident power (P) is scanned from 20 μW to 2 W, at several temperatures (T), with no biasing dc field H_0. As evidenced from the plots the microwave response is linear in the incident power at temperatures above T_c but is highly nonlinear below T_c. Actually, the nonlinearity manifests at quite low power levels (a few mW) as shown in the inset of Fig. 2. Notice that the plots were produced in a xy recorder with the y input fed from a diode detector in the heterodyne receiver and the x axis fed directly from

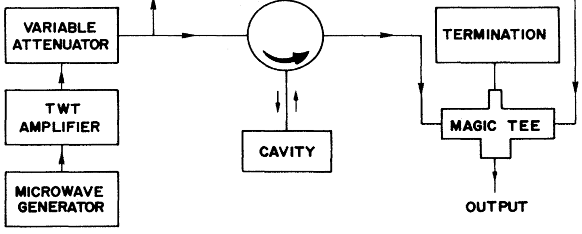

FIG. 1. Schematic experimental arrangement for measurements of the nonlinear microwave response of Y-Ba-Cu-O.

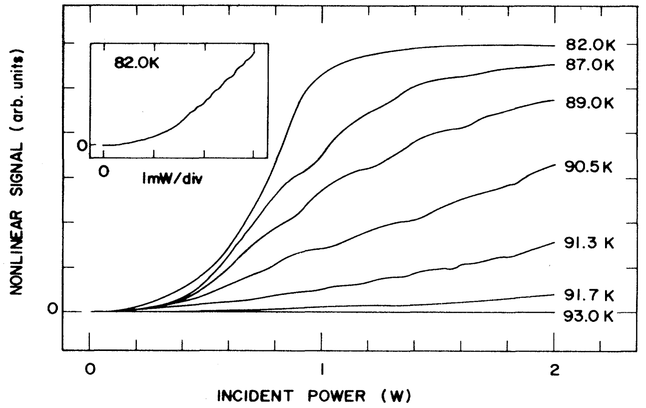

FIG. 2. Power dependence of the nonlinear (cw) microwave response of Y-Ba-Cu-O at 9.4 GHz (cavity $Q=2000$) for several temperatures. Inset shows the onset of the nonlinear response at low power levels measured with sensitivity 10^3 larger than in the main figure. It is worth noting that below 200 mW the effect is the same whether cw or pulsed microwave excitation is used.

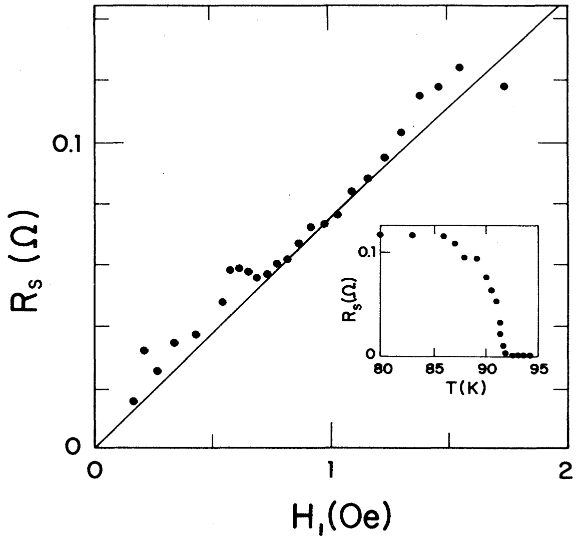

FIG. 3. Circles: surface-resistance data at $T=80$ K and $H_0=0$ vs microwave magnetic field amplitude. Solid line: fit with Eq. (3) of text. Inset shows R_s as a function of temperature in the vicinity of T_c with incident power fixed at 2 W. The data were obtained with pulsed (width: 50 μs; rate: 500 Hz) microwave to avoid heating effects at higher power levels.

a power meter. The continuous-wave (cw) microwave power was scanned by manually rotating the attenuator knob. These plots are shown here for a clear demonstration of the nonlinear response. However they were not used for quantitative measurements because the signal from the diode is not linear in the microwave power and also because there is some sample heating with cw power. The heating is more pronounced for $P > 500$ mW, particularly at temperatures close to T_c, and combined with the irregular power scan rate is responsible for the wiggles in the data of Fig. 2. It is worth noting that the nonlinear response below 200 mW is the same whether cw or pulsed microwave (width 50 μs, 500 pulses per second) is used, indicating that heating is negligible at lower power levels.

The nonlinear signal of the microwave bridge arises essentially from changes in the amplitude of the cavity reflection coefficient with power, since the phase change was found to be negligible. Thus we can determine the dependence of the sample surface resistance on H_0 and H_1 by measuring the reflection coefficient $\Gamma(H_0,H_1)$ directly from the variation of the reflected power P_r with P. This was done by using the calibrated attenuator to determine P and a power meter at the bridge output to measure variations in P_r. The surface resistance was determined with the cavity perturbation expression $R_s=(4\pi\omega V_c/c^2AQ)\Gamma$ where ω is the microwave frequency, V_c is the cavity volume, c is the speed of light, and A the sample surface area.

Figure 3 shows measurements of the surface resistance at $H_0=0$. The main figure represents the variation of R_s with the microwave field H_1 at $T=80$ K. As expected, at small microwave power the resistance vanishes at $H_0=0$. However the resistance increases with power implying that microwave loss exists even in the absence of a dc field. From the data of Fig. 3 we see that $R_s(H_1)$ has a linear dependence on H_1 with slope 0.07 Ω/Oe. The inset shows the temperature dependence of R_s at $P=2W$ ($H_1\simeq1.85$ Oe), demonstrating clearly the superconducting nature of the nonlinear response.

Finally, in Fig. 4, we compare the dependence of the reflected signal (a) with microwave power at $H_0=0$ and (b) with H_0 at constant power (20 mW) at $T=80$ K. The

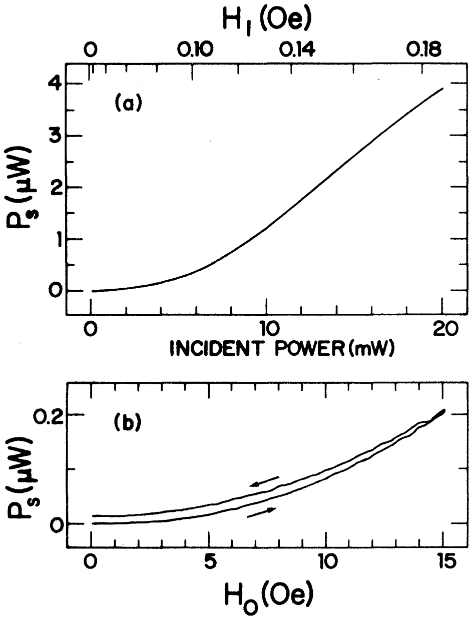

FIG. 4. Measured bridge output power P_s as a function of (a) incident (cw) power and (b) static magnetic field with incident power fixed at 20 mW. The change in the power reflected by the cavity is approximately $2P_s$. At this power level there is no observable heating effect.

signal in (b), which shows the usual hysteresis effect,[10–13] was obtained with the field H_0 applied parallel to the microwave field H_1. However, a nearly identical result is obtained if $\mathbf{H}_0$ is perpendicular to $\mathbf{H}_1$. A striking feature of the data in Fig. 4 is the fact that the signal obtained with the variation of H_1 (20 mW corresponds to $H_1=0.185$ Oe) is much larger than the one resulting from a corresponding variation of H_0 (at constant H_1). This result is confirmed by the fact that the nonlinear response data of Fig. 2 is not altered by the application of dc fields up to 15 Oe. This shows that the microwave absorption of granular high-T_c superconductors is intrinsically nonlinear. From the data of Fig. 4(b) we find that the change in the surface resistance with H_0 is 0.0025 Ω/Oe, a value very similar to the one obtained in Ref. 10 and which is 28 times smaller than the slope of $R_s(H_1)$.

DISCUSSION

The microwave response of the high-T_c ceramic oxides has been interpreted in terms of several microscopic models. However, the only one for which there is quantitative comparison between theory and experiment is the model of Portis *et al.*[10] based on the interaction between the microwave currents and free or pinned fluxons created by an external magnetic field. Therefore, we discuss our results in terms of that model. Due to the granular nature of the ceramic superconductors, the critical field H_{c1}^* for the entry of fluxons can be as small as 50 mOe.[6] According to the model of Portis *et al.*,[10] fluxons created by a biasing dc field $H_0 > H_{c1}^*$ with density $\mu f H_0/\phi_0$ (ϕ_0 is the flux quantum, μ is the permeability, and f is a proportionality constant of order 0.1) interact with the microwave current $\mathbf{J}$ with a force $(\phi_0/c)(\hat{\mathbf{z}}\times\mathbf{J})$, where $\hat{\mathbf{z}}$ is the unit vector in the direction of $\mathbf{H}_0$. This results in fluxon motion with the microwave frequency with velocity $\mathbf{v}=(\phi_0/c\eta)(\hat{\mathbf{z}}\times\mathbf{J})$, where η is the fluxon viscous damping constant. The fluxons in motion induce a microwave electric field that opposes the current, giving rise to absorption and dispersion of the incoming radiation. The interaction between the sample and the microwave radiation can be expressed in terms of a surface impedance, whose real part is the surface resistance[10]

$$R_s(H_0)=X_0[-1+(1+aH_0^2)^{1/2}]^{1/2}/\sqrt{2}\,, \tag{1}$$

where $X_0=4\pi\omega\mu\lambda_L/c^2$ is the surface reactance in zero field, λ_L is the London penetration length, and

$$a=(f\phi_0/4\pi\omega\eta\lambda_L^2)^2\,. \tag{2}$$

The experimental value of 0.0025 Ω/Oe obtained for $R_s(H_0)$ is consistent with this model if we use parameters in the range quoted in Ref. 10, namely, $\mu f \simeq 0.1$, $\eta \sim 10^{-9}$ cgs units, and $\lambda_L \simeq 300$ Å.

In order to use the same model to explain the observed nonlinear response, we make the assumption that with $H_0=0$ fluxons are pushed in and out of the sample by the rf magnetic field when it exceeds H_{c1}^* during part of the cycle. For $H_{c1}^* \sim 50$ mOe, this occurs at power levels as low as 0.8 mW in a cavity with $Q=2000$. This results in a fluxon density proportional to the rf magnetic field and, consequently, the microwave absorption is nonlinear in the microwave intensity. Assuming that $H_1 \gg H_{c1}^*$ and $H_0=0$, the model gives a surface resistance which depends on the rf field as

$$R_s(H_1)=X_0[-1+(1+aH_1^2/2)^{1/2}]^{1/2}/\sqrt{2}\,. \tag{3}$$

For $aH_1^2 \ll 1$ this gives the observed linear dependence of $R_s(H_1)$. The solid line in Fig. 3 represents a fit of Eq. (3) to the data with $X_0a^{1/2} \simeq 0.14$ Ω/Oe, a value much larger than the one used to fit the $R_s(H_0)$ data. At present, we do not have a plausible argument to explain this discrepancy. We note, however, that a similar difficulty is encountered if one tries to interpret the microwave absorption in terms of Josephson junction currents. According to Ref. 15, the dependence of the junction loop current on H_0 is of the same magnitude as on H_1, contrary to our observations.

In summary, we have shown that the microwave absorption of ceramic Y-Ba-Cu-O in the superconducting phase is highly nonlinear in the microwave power. Contrary to general belief, at microwave power levels above 1 mW in a cavity with $Q > 2000$, the microwave loss in the absence of a dc biasing field is not negligible. This explains the surprisingly large absorption previously observed[4] below the superconducting transition. This result raises difficulties for technological applications of high-T_c ceramic oxides at high frequencies.

ACKNOWLEDGMENTS

We gratefully acknowledge Professor F. Waldner and Professor A. Portis for introducing us to the microwave properties of superconductors in lectures and discussions at the Physics Institute, University of Zurich, L. Pauli for providing the samples used in the experiments, M. Macedo for helping in some measurements, and A. V. Nascimento for technical support. We also thank Professor Portis for a critical reading of the first version of the paper and for offering several suggestions. This work has been supported by Financiadora de Estudos e Projetos, Conselho Nacional de Desenvolvimento Científico e Tecnológico e Coordenação de Aperfeiçoamento do Pessoal de Ensino Superior.

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[1] W. P. Beyermann, B. Alavi, and G. Grüner, Phys. Rev. B **35**, 8826 (1987).

[2] S. Sridhar, C. A. Shiffman, and H. Hamdeh, Phys. Rev. B **36**, 2301 (1987).

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

[4] M. Poirier, G. Quirion, K. R. Poeppelmeier, and J. P. Thiel, Phys. Rev. B **36**, 3906 (1987).

[5] C. Rettori, D. Davidov, I. Belaish, and I. Felner, Phys. Rev. B **36**, 4028 (1987).

[6] A. Raboutou, P. Peyral, J. Rosenblatt, C. Lebeau, O. Pena, A. Perrin, C. Perrin, and M. Sergent, Europhys. Lett. **4**, 1321 (1987).

[7] K. W. Blazey, K. A. Müller, J. G. Bednorz, W. Berlinger, G. Amoretti, E. Buluggin, A. Vera, and F. C. Matacotta, Phys. Rev. B **36**, 7241 (1987).

[8] K. Khachaturyan, E. R. Weber, P. Tejedor, A. M. Stacy, and A. M. Portis, Phys. Rev. B **36**, 8309 (1987).

[9] K. W. Blazey, A. M. Portis, and J. G. Bednorz, Solid State Commun. **65**, 1153 (1988).

[10] A. M. Portis, K. W. Blazey, K. A. Müller, and J. G. Bednorz, Europhys. Lett. **5**, 467 (1988).

[11] M. Stalder, G. Stefanicki, M. Warden, A. M. Portis, and F. Waldner, Physica C **153–155**, 659 (1988).

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

[13] S. H. Glarum, J. H. Marshall, and L. F. Schneemeyer, Phys. Rev. B **37**, 7491 (1988).

[14] W. J. Tomasch, H. A. Blackstead, S. T. Ruggiero, P. J. McGinn, J. R. Clem, K. Shen, J. W. Weber, and D. Boyne, Phys. Rev. B **37**, 9864 (1988).

[15] C. Jeffries, Q. H. Lam, Y. Kim, L. C. Bourne, and A. Zettl, Phys. Rev. B **37**, 9840 (1988).

[16] O. V. Abramov, G. I. Leviev, V. G. Pogosov, and M. R. Trunn, Pis'ma Zh. Eksp. Teor. Fiz. **46**, 433 (1987) [JETP Lett. **46**, 546 (1987)].

[17] I. Ciccarello, M. Guccione, M. Li Vigni, and A. Sarro, Europhys. Lett. **7**, 185 (1988).