Observation of the Nonlinear Meissner Effect in YBCO Thin Films: Evidence for a d-Wave Order Parameter in the Bulk of the Cuprate Superconductors

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We present experimental evidence for the observation of the nonlinear Meissner effect in high-quality epitaxial yttrium barium copper oxide thin films by measuring their intermodulation distortion at microwave frequencies versus temperature. Most of the films measured show a characteristic increase in nonlinearity at low temperatures as predicted by the nonlinear Meissner effect. We could measure the nonlinear Meissner effect because intermodulation distortion measurements are an extremely sensitive method that can detect changes in the penetration depth of the order of 1 part in 10^5.

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The nonlinear Meissner effect (NLME) was predicted by Yip and Sauls [1,2] in superconductors with d-wave symmetry. Dahm and Scalapino (DS) [3,4] applied the NLME to the nonlinear microwave properties of the high-temperature superconductors with emphasis on harmonic generation and intermodulation distortion (IMD). They predicted that the London penetration depth depends on the magnetic field in such a way that, as the temperature is lowered, the nonlinearity increases. Previous attempts to observe the NLME by means other than microwave measurements have yielded either negative or ambiguous results [5]. A previous report [6] did show an indication of an increase in microwave-frequency IMD at low temperatures. We report here measurements from which we extract the nonlinear penetration depth that exhibits both the predicted field and temperature dependence. The measurements were carried out on yttrium barium copper oxide (YBCO) films using a microwave-frequency stripline-resonator technique that characterizes the nonlinearity by intermodulation distortion. In this procedure, two closely spaced tones at frequencies f_1 and f_2 are applied to the resonator. The third-order mixing products at frequencies $2f_1 - f_2$ and $2f_2 - f_1$ are then measured. The strength of the IMD signals can be directly related to the nonlinear penetration depth.

In the NLME, the penetration depth λ is given by [3,4]

$$\lambda(T, j) = \lambda(T)\left[1 + b(T)\left(\frac{j}{j_c}\right)^2\right], \quad (1)$$

where $b(T)$ is an angle-averaged coefficient that diverges as $1/T$ at low-temperature, j is the microwave current, and j_c is the depairing critical current. DS suggest using a value of 3×10^8 A/cm^2 for j_c in YBCO. The j^2 dependence in Eq. (1) applies at low field and changes over to $|j|$ at a crossover value as discussed in [3]. The angle averaging to obtain $b(T)$ accounts for the different directions of current flow in the meandering stripline with respect to the material coordinates. However, b does not vary strongly with direction of current flow.

As first proposed by DS, measurements of either the IMD or third harmonic generation should show a temperature and field dependence given by the NLME. We show that measurements of the IMD in microwave resonators are ideal for the observation of the NLME since they are a very sensitive characterization of the nonlinearity.

Recently, the question of the difference between the surface and bulk properties of the high-temperature superconductor materials has been discussed in the literature [7]. The majority of reports support the existence of d-wave symmetry in the bulk and coexistence of d- and is-wave components within a coherence length ξ_0 of the surface. The microwave experiments reported here are definitely probes of the bulk, since the penetration of the microwave fields is governed by $\lambda \gg \xi_0$. Our results suggest strongly that a d-wave component is present in the bulk.

To relate the measured quantities to the nonlinear penetration depth, we follow DS [3,4] and assume that the microwave-frequency is small compared with the reciprocal of the quasiparticle relaxation time. In order to simplify the expressions for the IMD, we rewrite Eq. (1) as

$$\lambda(T, j) = \lambda(T) + \lambda_2 j^2, \quad (2)$$

where $\lambda_2 = \lambda_0 b(T)/j_c^2$.

Referring to the equivalent circuit shown in Fig. 1, the inductance L depends on λ [8,9]. If one assumes that the nonlinear inductance is given by $L = L_0 + L_2 I^2$, where L_2 is a constant proportional to λ_2 [3,4] and I is the total rf current in the resonator, the expression for the IMD power P_{IMD} is [4]

$$P_{\mathrm{IMD}} = \left(\frac{4}{\pi}\right)^2 \omega^2 L_2^2 \frac{[2r_v(1 - r_v)]^4 Q_c^4 P^3}{\pi^2 Z_0^4}, \quad (3)$$

where ω is the angular frequency, r_v is the voltage in-

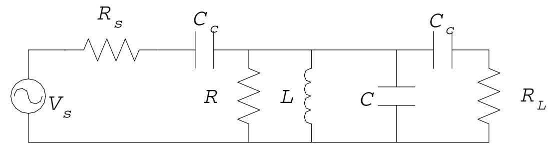

FIG. 1. Equivalent circuit of a resonator. R, L, and C are given by the transmission line parameters [9]. C_c is the coupling capacitor, and $R_s = R_L = 50\ \Omega$ are the load and source impedances.

sertion ratio, related to the insertion loss IL in decibels (dBm) by $IL = -20 \log r_v$, Q_c is the unloaded Q of the resonator, P is the input power, and Z_0 is the characteristic impedance of the resonator.

It is also most illustrative to plot the IMD results versus the circulating power in the resonator, given by

$$P_{\rm circ} = \frac{4r_v(1 - r_v)Q_c P}{\pi}, \tag{4}$$

and then $P_{\rm norm}$, the normalized IMD power that is used to compare the data from various resonators is

$$P_{\rm norm} = \frac{P_{\rm IMD}}{r_v(1 - r_v)Q_c}. \tag{5}$$

The $P_{\rm norm}$ is proportional to L_2^2 and, therefore λ_2^2, and removes the effects of different Q and r_v.

The IMD measurements reported here have been carried out on six films from various sources. All of the films were epitaxial c-axis normal YBCO and some had dopants added during the growth process. Table I gives the details of the films used [10–12].

The films were patterned using standard photolithography and wet etching, assembled with ground planes to form stripline resonators, and measured by a technique, described previously [13,14], in which the Q and resonant frequency f_0 of the resonator are measured as a function of the microwave power at temperatures between 1.7 K and T_c. The results are converted into the effective surface resistance $R_S(I_{\rm rf})$ and reactance $X_S(I_{\rm rf})$ where $I_{\rm rf}$ is the microwave current. Although it is well known that the current density at the edges is strongly enhanced, we have previously demonstrated that etching does not affect the nonlinear surface impedance [15]. At the low powers of interest for the NLME, and for the measurements reported here, the j at the edge remains below j_c, and the edges remain in the Meissner state. The measurements were done at the fundamental frequency of 1.5 GHz for the case of the $LaAlO_3$ substrates and 2.3 GHz for the Al_2O_3 substrate. These are all high-quality films with resonator Q values of order 10^5 and very low R_S. Results of measurements of $Z_S(I_{\rm rf})$ for the pulsed laser deposition (PLD) films on $LaAlO_3$ have been reported previously [16].

The same resonators used for the Z_S measurements are used for the IMD measurements. Figure 2 shows the measurements of the normalized IMD power as a function of the circulating power in the resonator for the pure $YBa_2Cu_3O_{7-\delta}$ film, number 1 in Table I, for several temperatures. The circulating power is given in dBm, defined as $10 \log(P_{\rm circ}/1\ {\rm mW})$. Likewise, the ordinate is in dBm from Eq. (5). For clarity, curves for only four temperatures are shown. The simple ansatz of Eq. (2) predicts $P_{\rm IMD} \sim P_{\rm circ}^3$, yielding a slope of three. A dashed line in Fig. 2 with a slope of three is included as a reference. At low powers, the dependence is slope three, indicating a quadratic dependence of λ on current. At intermediate powers, the slope changes over to a value less than two for the lowest temperatures. A slope of two indicates a dependence of λ on the modulus of the current in agreement with the prediction of the NLME. For temperatures below approximately 70 K, the curves merge near 20 dBm circulating power. The slope-two behavior is an indication that the simple ansatz of Eq. (2) no longer holds and the behavior instead is $\lambda = \lambda_0 + \lambda_2|I|$. Subsequently, at the highest powers the slope becomes three again. Note that at the lowest powers the IMD *increases* as the temperature decreases, in agreement with the predictions of the NLME. At powers higher than the intersection point the slope becomes three once again, and the IMD is monotonically increasing with increasing temperature. Above approximately 70 K, the IMD shows a simple slope-three behavior. The behavior in Fig. 2 was shown by most of the films measured but was clearest for the film shown.

Figure 3 shows the temperature dependence of the IMD power in Fig. 2 taken at constant circulating powers of 5, 20, and 40 dBm, corresponding to: the lowest for which good signal to noise ratio is obtained, the point where the curves cross in Fig. 2, and the highest power measured, respectively. At low power, an upturn at low temperature is evident. At 20 dBm circulating power, the curves are independent of temperature below 60 K, within experi-

TABLE I. Details of the films used in IMD measurements.

Number	Film type	Thickness (μm)	Substrate	Deposition method	Source
1	$YBa_2Cu_3O_{7-\delta}$	0.4	(100)$LaAlO_3$	Pulsed laser deposition (PLD)	Koren *et al.* [10]
2	$YBa_2Ni_{0.06}Cu_{2.94}O_{7-\delta}$	0.4	$LaAlO_3$	PLD	Koren *et al.*[10]
3	$YBa_2Zn_{0.06}Cu_{2.94}O_{7-\delta}$	0.4	$LaAlO_3$	PLD	Koren *et al.* [10]
4	$Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7-\delta}$	0.4	$LaAlO_3$	PLD	Koren *et al.* [10]
5	$YBa_2Cu_3O_{7-\delta}$	0.3	$LaAlO_3$	Sputtered	Anderson *et al.* [11]
6	$YBa_2Cu_3O_{7-\delta}$	0.23	r-plane-Al_2O_3	PLD	Lorenz *et al.* [12]

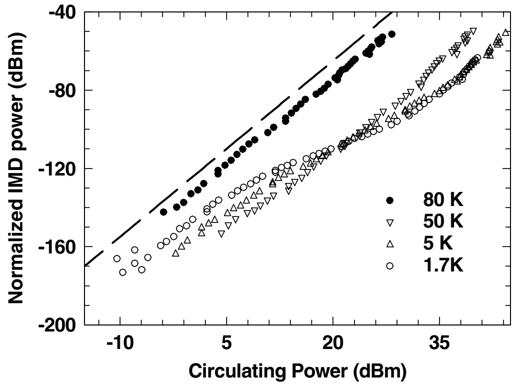

FIG. 2. IMD versus circulating power for selected temperatures. This is for film number 1, the PLD film of pure $YBa_2Cu_3O_{7-\delta}$. The dashed line illustrates slope 3, which is an indication of a P_{circ}^3 dependence. See Eqs. (3) and (4).

mental uncertainty. At high power, a decrease in IMD is observed at low temperature. At 5 dBm circulating power, the peak of the current density at the edge of the stripline is approximately $2.8 \times 10^5\ A/cm^2$, 2 orders of magnitude below the critical current density at low temperature. The peak magnetic field is about 4 Oe.

Figure 4 shows the temperature dependence of the measured IMD at 5 dBm circulating power for the films in Table I. Plotted is the IMD, scaled as described by Eq. (5), versus the reduced temperature. To obtain this data, each of the films was characterized like the pure, PLD YBCO film shown in Fig. 2. The actual range of temperature is from 1.7 to 85 K.

The film with the lowest IMD at the middle temperature range, which is thus the highest quality film, the pure PLD YBCO, number 1 in Table I, shows a significant upturn at low temperatures. This is the film shown in Figs. 2 and 3. The other films show either a smaller upturn or none at all in the case of the YBCO film on sapphire and the heavily Ca doped film. The convergence at low temperatures might be explained by postulating that the IMD is the sum of extrinsic and intrinsic contributions. If the extrinsic contribution dominates at intermediate tem-

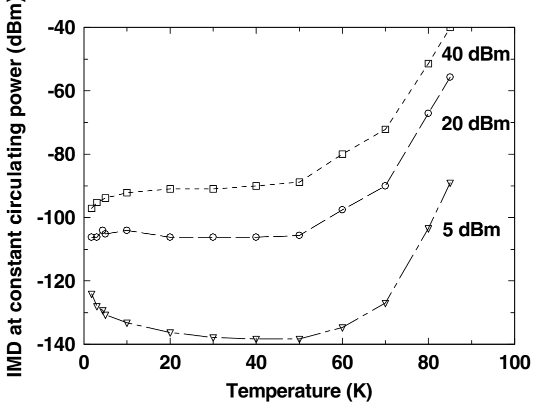

FIG. 3. IMD at constant circulating power versus temperature for the powers indicated.

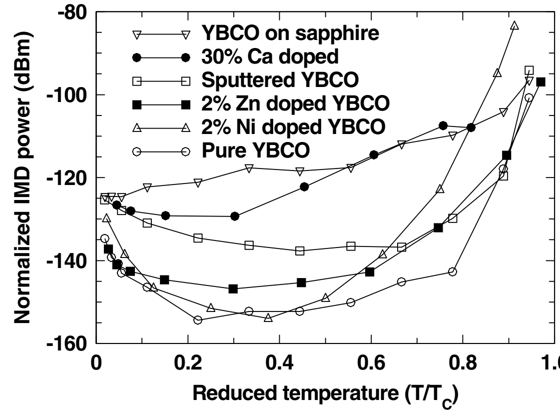

FIG. 4. Measurements of the IMD for the various films listed in Table I: empty circles: pure YBCO (number 1); empty triangles: Ni-doped, (number 2); filled squares: Zn doped, (number 3); filled circles: Ca doped, (number 4); empty squares: sputtered YBCO, (number 5), all on $LaAlO_3$; empty inverted triangles: YBCO on sapphire, (number 6).

peratures and tends to a constant value at low temperatures, which might be expected for weak links, for example, then at low enough temperatures the intrinsic IMD would eventually become larger than the extrinsic values. Films on sapphire are known to have more defects, and, in particular, microcracks, than YBCO films on other substrates. The sputtered film, number 5 in Table I, also was judged to be of lower quality than the best sputtered films due to high Z_S values. Not shown in Fig. 5 are further results from a different source, one on sapphire and one on $LaAlO_3$ that reproduce almost exactly these results, in that the film on sapphire shows much larger IMD at all temperatures than the YBCO film and shows no increase at low temperature while the YBCO film does.

To verify our results on YBCO, comparable measurements were made on a resonator of the same design fabricated from a sputtered niobium film [17] on a $LaAlO_3$ substrate. It is predicted [3,4] that s-wave superconductors show an IMD power decreasing exponentially at low temperatures because of the finite energy gap. The

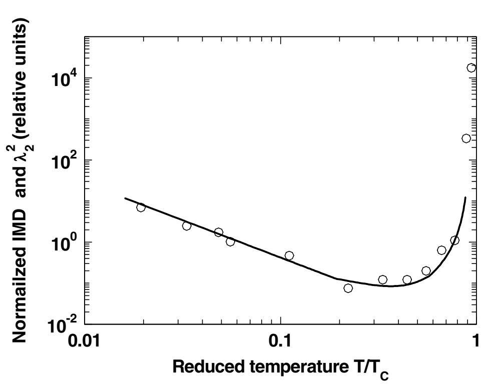

FIG. 5. Comparison of data and theory. The empty circles are the IMD data from film number 1. The solid line is the calculation of d-wave theory.

IMD data versus temperature (not shown) indicate that niobium does not show an upturn and becomes independent of temperature at low T as might be expected if it is limited by extrinsic effects. However, the measurements on niobium do not reach the same reduced temperature as YBCO because of the lower T_c and, thus, cannot show unambiguously the expected s-wave behavior. The most important result of the niobium IMD measurements is that the temperature-independent IMD response rules out any substrate effects in the observed increase of IMD in YBCO at low temperatures used.

DS [4] have calculated b in Eq. (1) for a d-wave superconductor with parameters appropriate for YBCO. The value of b depends upon the direction of current flow. We have used an angle-averaged value here because in our meanderline resonator all directions are sampled. Moreover, the variation is relatively small, the shape of the curve remains similar, and the temperature dependence shows the same low-temperature $1/T^2$ divergence. To compare with the experiment, we have plotted in Fig. 5 λ_2^2 from the DS calculation together with the measured data from film number 1 of Table I. This is the data at 5 dBm circulating power in Fig. 3. We have plotted relative values only. The magnitude of the curves has been taken as a free parameter. The temperature dependence of the measured data agrees well with the calculations. Especially note the low-temperature behavior that in both data and calculation shows very accurately a $1/T^2$ divergence. It is obvious that the other films depicted in Fig. 4 do not agree as well with the theory due probably to extrinsic effects.

Taking the absolute magnitude of the calculated IMD in Fig. 5 as a free parameter is equivalent to the use of j_c, the depairing critical current of Eq. (1), as a free parameter. We have made an estimate of the value of j_c for the pure YBCO, film number 1, needed to give agreement in absolute value. Details of this procedure will be reported elsewhere. Our value thus obtained is 4.8×10^8 A/cm^2, which compares well with the value of 3×10^8 suggested by DS. Since the IMD level is proportional to $1/j_c^4$ as follows from Eq. (3) [4], the j_c values for the other films in Fig. 4, except for the Ni-doped film, will be considerably lower if one assumes that they can be described by the DS model. Clearly, however, the DS model does not hold for films that show a monotonically decreasing dependence of IMD on temperature. Other effects such as weak links seem to be contributing to the IMD for these films. It may be that the IMD in the other films results from a sum of the intrinsic IMD given by the DS model and IMD generated by the effects of weak links or other defects.

In conclusion, strong experimental evidence is given for the observation of the nonlinear Meissner effect in high-quality epitaxial thin films of YBCO by measuring their IMD at microwave frequencies and low temperatures. The best film presented here exhibits the predicted $1/T^2$ dependence of the IMD at low power and low-temperature. Defects, impurities, and perhaps other extrinsic effects in some of the films limit the observability of the NLME, which is an intrinsic property. The magnitude of the predicted IMD is within a factor of 3 of the predicted effect and can be fit with a reasonable adjustment of the depairing critical current by about 1.7 from that proposed [3,4]. We could measure the NLME because IMD measurements are an extremely sensitive method that can detect changes in the penetration depth of the order of 1 part in 10^5 [18].

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