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 \(10^5\).

We 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. 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 \(\lambda\) is given by [3,4]

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

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 \(3 \times 10^8\) 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 \(\xi (0)\) of the surface. The microwave experiments reported here are definitely probes of the bulk, since the penetration of the microwave fields is governed by \(\lambda > \xi (0)\). 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

\[
\lambda(T, j) = \lambda(T) + \lambda_2 j^2,
\]

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

Referring to the equivalent circuit shown in Fig. 1, the inductance \(L\) depends on \(\lambda\) [8,9]. If one assumes that the nonlinear inductance is given by \(L = L_0 + L_2 j^2\), where \(L_2\) is a constant proportional to \(\lambda_2\) [3,4] and \(I\) is the total rf current in the resonator, the expression for the IMD power \(P_{IMD}\) is [4]

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

where \(\omega\) is the angular frequency, \(r_v\) is the voltage in-
insertion ratio, related to the insertion loss $IL$ in decibels (dBm) by $IL = -20 \log_{10} P_c$, $Q_c$ 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

$$P_{\text{circ}} = \frac{4r_v(1-r_v)Q_cP}{\pi},$$

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

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

The $P_{\text{norm}}$ is proportional to $L_2$ and, therefore $\lambda_2$, 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 $R_S(I_{\text{rf}})$ and reactance $X_S(I_{\text{rf}})$ where $I_{\text{rf}}$ 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$_2$O$_3$ substrate. These are all high-quality films with resonator $Q$ values of order $10^5$ and very low $R_S$.

Results of measurements of $Z_S(I_{\text{rf}})$ 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$_2$Cu$_3$O$_{7-\delta}$ film, number 1 in Table I, for several temperatures. The circulating power is given in dBm, defined as $10\log(P_{\text{circ}}/1\text{ mW})$. 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 $P_{\text{IMD}} \sim P_{\text{circ}}^3$, 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 $\lambda$ 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 $\lambda$ 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-

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

<table>
<thead>
<tr>
<th>Number</th>
<th>Film type</th>
<th>Thickness ($\mu$m)</th>
<th>Substrate</th>
<th>Deposition method</th>
<th>Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>YBa$_2$Cu$<em>3$O$</em>{7-\delta}$</td>
<td>0.4</td>
<td>(100)LaAlO$_3$</td>
<td>Pulsed laser deposition (PLD)</td>
<td>Koren et al. [10]</td>
</tr>
<tr>
<td>2</td>
<td>YBa$<em>2$Ni$</em>{0.66}$Cu$<em>{2.94}$O$</em>{7-\delta}$</td>
<td>0.4</td>
<td>LaAlO$_3$</td>
<td>PLD</td>
<td>Koren et al. [10]</td>
</tr>
<tr>
<td>3</td>
<td>YBa$<em>2$Zn$</em>{0.66}$Cu$<em>{2.94}$O$</em>{7-\delta}$</td>
<td>0.4</td>
<td>LaAlO$_3$</td>
<td>PLD</td>
<td>Koren et al. [10]</td>
</tr>
<tr>
<td>4</td>
<td>Y$<em>{0.7}$Cu$</em>{0.3}$Ba$_2$Cu$<em>3$O$</em>{7-\delta}$</td>
<td>0.4</td>
<td>LaAlO$_3$</td>
<td>PLD</td>
<td>Koren et al. [10]</td>
</tr>
<tr>
<td>5</td>
<td>YBa$_2$Cu$<em>3$O$</em>{7-\delta}$</td>
<td>0.3</td>
<td>LaAlO$_3$</td>
<td>Sputtered</td>
<td>Anderson et al. [11]</td>
</tr>
<tr>
<td>6</td>
<td>YBa$_2$Cu$<em>3$O$</em>{7-\delta}$</td>
<td>0.23</td>
<td>r-plane-Al$_2$O$_3$</td>
<td>PLD</td>
<td>Lorenz et al. [12]</td>
</tr>
</tbody>
</table>
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 $2.8 \times 10^5 \, \text{A/cm}^2$, 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 temperatures 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 $\text{LaAlO}_3$ 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 $\text{LaAlO}_3$ 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
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 $\lambda^2$ 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 the temperature-independent IMD response rules out any substrate effects in the observed increase of IMD in YBCO at low temperatures used.

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 observed and can be fit with a reasonable adjustment of the depairing critical current by about 1.7 from that proposed [3,4].

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[10] G. Koren, A. Gupta, R. J. Beserman, M. I. Lutwyche, and R. B. Laibowitz, Appl. Phys. Lett. 55, 2450 (1989). The high-quality epitaxial PLD films used in the present study were grown using the 355 nm laser wavelength. They have a room temperature resistivity of 0.2 m$\Omega$ cm, $T_c = 90-91$ K, and $d(\rho)/dT = -4.5 \times 10^8$ A/cm$^2$.