Disorder effects on the intrinsic nonlinear current density in YBa$_2$Cu$_3$O$_{7-\delta}$

Brian M. Andersen, James C. Booth, and P. J. Hirschfeld

1 Department of Physics, University of Florida, Gainesville, Florida 32611-8440, USA
2 National Institute of Standards and Technology, Boulder, Colorado 80303, USA

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We present harmonic generation measurements of the intrinsic nonlinear current density $j_2$ of YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO) films at temperatures close to the $T_c$. Experiments on a range of different quality samples allow us to extract the dependence of $j_2$ on the penetration depth of the superconductor. In order to model these results, we calculate the intrinsic nonlinear current response of $d_{2-2}$-wave superconductors in the Meissner regime in the presence of nonmagnetic impurities within the self-consistent T-matrix approximation.

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Introduction

Superconductors with $d$-wave pairing symmetry should exhibit an unusual angle dependent nonlinear Meissner effect. This has the effect of significantly altering the current density dependence of the superfluid density at low temperatures, and dramatically affects the nonlinear response of superconductor transmission lines or resonators. Recently, intermodulation measurements of resonators fabricated from epitaxial YBCO thin films revealed a sharp increase in the observed nonlinearity at low temperatures, a result that was originally predicted by Dahm and Scalapino. While the measured temperature dependence suggests that this observed nonlinear response has intrinsic origins, the effect of impurities on the nonlinear behavior in high $T_c$ superconductors has not been explored systematically by experiments. This issue is important since the nonlinear response and its unwanted intermodulation products can severely limit the applications of superconducting devices used for e.g. communication filters.

In this paper we report on measurements of the nonlinear current density scale $j_2$ determined from third-harmonic generation experiments for a range of YBCO thin-film samples at a fixed temperature (76 K), but which display varying levels of growth-induced defects, reflected in the measured penetration depth. The samples were fabricated under a variety of growth conditions, and showed variations in the penetration depth over the range from 0.25 - 0.4 µm at 76 K. Subsequent third-harmonic generation measurements on these samples allowed us to examine the nonlinear current density scale $j_2$ as a function of the measured penetration depth, in order to explore the effects of impurities on the nonlinear response. We compare these measured results with theoretical predictions for a $d_{2-2}$-wave superconductor in the Meissner regime in the presence of nonmagnetic impurities.

For a superconductor, the superfluid density is a function of the current density. This manifests itself experimentally as a dependence on current density of the superconductor magnetic penetration depth. The exact form of the current-dependent penetration depth depends on the nature of the superconductor, including the details of the superconductor energy gap. Since for most cases the current-density dependence of the penetration depth $\lambda(j)$ is very small, we can use a polynomial approximation for this quantity. We expect the change in the penetration depth to be an even function of the current density $j$, so the lowest-order term in this low-temperature expansion is quadratic:

$$\left(\frac{\lambda(j)}{\lambda_0}\right)^2 = 1 + \langle j/j_2 \rangle^2,$$

where $\lambda_0$ is the zero-current penetration depth and $j_2$ sets the scale of the nonlinear current density ($j_2$ is sometimes also referred to as the intermodulation critical current). In a $d$-wave superconductor, Eq. (1) is known to break down at low temperatures due to singular contributions to the current from nodal quasiparticles. The nonlinear current density scale $j_2$ characterizes the strength of the current-density dependence of the penetration depth, and its behavior as a function of e.g. temperature and disorder can reveal important details regarding the nature of the superconducting state. It is the measured temperature dependence of $j_2$ extracted from nonlinear microwave experiments that reveals the $d$-wave behavior predicted by Dahm and Scalapino. In the present work we examine the dependence of $j_2$ on the penetration depth in order to explore the role of disorder in the nonlinear response of high $T_c$ superconductors.

Experiment

Experimentally, there are a number of different ways to measure $\lambda(j)$. However, the large value for $j_2$ that results from intrinsic effects means that very large current densities are required to produce even very small changes in $\lambda(j)$. This means that extrinsic effects due to self-heating or vortex motion, for example, may easily mask the intrinsic behavior, making the extraction of reliable
values for $j_2$ extremely challenging. One technique that possesses the required sensitivity to small changes in $\lambda$ is the mutual inductance technique reported by Claassen et al.\textsuperscript{12}, which used a persistent current trapped in the thin-film samples to measure $\lambda(j)$ as a function of dc current. By carefully accounting for heating effects of the dc current induced in the measurement coils this technique was able to extract an accurate value for $j_2$. The experiments demonstrated the expected quadratic dependence of penetration depth on dc current density\textsuperscript{10}.

It has also been realized that a current-density dependent penetration depth will result in a current-dependent inductance per unit length for a superconducting thin-film transmission line\textsuperscript{8-11}, or a current-dependent film inductance for mutual inductance experiments\textsuperscript{12}. Such a nonlinear inductance will produce mixing or harmonic products for single-frequency or two-tone stimuli, which can be detected with remarkable sensitivity using straightforward spectrum analysis\textsuperscript{13} or lock-in techniques. Use of such harmonic mixing or harmonic generation techniques means that nonlinear effects generated by even a very small current-dependent contribution to the penetration depth can be detected fairly easily even for modest applied current densities. In what follows, $j_2$ determined from both third harmonic generation experiments at microwave frequencies, as well as from mutual inductance measurements at audio frequencies, is examined for YBCO samples that contain differing amounts of growth-induced defects, as reflected in the measured penetration depth.

For the microwave results described here, we use patterned coplanar waveguide transmission lines of different lengths and cross-sectional geometries fabricated from YBCO films with thicknesses in the range 50-400 nm. The YBCO thin films were grown by pulsed laser deposition, as well as by reactive co-evaporation. All of the films were grown on LaAlO\textsubscript{3} substrates, and all had superconducting transition temperatures in the 88-90 K range. Some films were intentionally grown under different growth conditions\textsuperscript{14} while others displayed differences that were simply the result of random process variations. For the microwave transmission line measurements, we apply a single-tone incident signal (typically 2-6 GHz), and measure the transmitted signal at the fundamental frequency as well as at the third harmonic frequency. From the measured third harmonic signal versus incident power, along with the transmission line dimensions and measured penetration depth, we can extract $j_2$\textsuperscript{12}. For transmission lines of different lengths and cross-sectional geometries fabricated from the same thin-film sample, we obtained consistent values\textsuperscript{13} for $j_2$. We also obtained consistent $j_2$ values from results of two-tone intermodulation measurements in transmission-line resonators, as well as from analysis of the power-dependent impedance derived from resonator measurements performed as a function of rf power\textsuperscript{13}. The self-consistency of the results for $j_2$ extracted from these many different measurements suggests that we are measuring an intrinsic, material-dependent nonlinear effect, as opposed to extrinsic nonlinear effects due to e.g. vortex motion or heating effects. The results of $j_2$ measurements at 76-77 K are shown for a number of different samples in Fig. 1 as a function of the measured penetration depth (also determined at 76-77 K).

As mentioned above, mutual inductance measurements at audio frequencies can also provide a measure of $j_2$\textsuperscript{12}. For these measurements, the unpatterned superconducting film is sandwiched between two coils driven at a frequency in the audio range (typically 10 kHz). Such an experimental arrangement can be used to derive $j_2$ in two different ways: (1) by measuring changes in the penetration depth as a function of a dc current induced in the superconducting film, or (2) by measuring the signal generated at the third harmonic of the drive signal using a lock-in amplifier. The two approaches have been shown\textsuperscript{12} to give consistent results for $j_2$ at 76 K, and $j_2$ measured by the former procedure has been shown to agree quantitatively with the third-harmonic generation results at microwave frequencies\textsuperscript{10}. The results of the mutual inductance determination of $j_2$ for a variety of samples from Ref.\textsuperscript{12} are also plotted in Fig. 1 as a function of the measured penetration depth. The agreement demonstrated between the microwave-frequency harmonic generation measurements and the mutual inductance techniques for $j_2$ values further supports our assertion that the nonlinear effects quantified by $j_2$ are intrinsic in nature. For the films measured by the mutual inductance technique, the penetration depth was measured also by

![FIG. 1: Intrinsic nonlinear current density scale $j_2$ versus penetration depth $\lambda$ extracted from a range of YBCO samples of different quality. Here we show the results obtained from the measured third harmonic signals ($IP_3$) at 76 K and the mutual inductance (MI) results by Claassen\textsuperscript{12} at 77 K.](image-url)
quasiparticles: the density of states for comoving and countermoving rate proportional to the impurity concentration where $\Gamma = n \cot \delta_0$, where $\delta_0$ is the s-wave scattering phase shift and $N_0$ is the density of states at the Fermi level. The strong scattering limit (unitary limit) is characterized by $c = 0$, whereas for the Born limit $c \gg 1$. For simplicity we have assumed a circular Fermi surface and study current flow along the nodal direction. The $d_{x^2-y^2}$-wave order parameter relevant for the high-$T_c$ cuprate materials can be written as

$$\Delta(\Theta) = \Delta_0 \sin(2\Theta).$$

We have included the suppression of $T_c$ and $\Delta_0$ by solving Eq. (9) and (10) together with the self-consistent gap equation

$$\frac{1}{g} = 2\pi T \sum_n \int_0^{\pi \omega_n} d\omega \frac{\sin^2(\omega)}{\omega_n^2 + \Delta^2(\Theta)},$$

where $g$ denotes the coupling constant and the sum is restricted by an upper energy cut-off $\omega_c$.

The superfluid density $n_s(j_s, T)$ is defined from the relation

$$j = \frac{n_s(j_s, T)}{n} j_s,$$

and in the local approximation, is related to the London penetration depth $\lambda(j_s, T)$ by

$$\left( \frac{\lambda(j_s, T)}{\lambda_0} \right)^2 = \frac{n}{n_s(j_s, T)},$$

where $n$ is the total electron density and $\lambda_0$ denotes the zero-current penetration depth. In agreement with Eq. (11) and (12), it is through the expansion of the superfluid density $n_s$ in terms of $j_s$ that $j_2$ is defined by the following relation

$$n_s(j_s, T) = n_s(T) \left[ 1 - \left( \frac{j_s}{j_2} \right)^2 \right].$$

Here, $j_2$ can be expressed as

$$\left( \frac{j_2}{j_c} \right)^2 = \frac{n_s(T)/n^3}{\beta(T)},$$

where

$$\frac{n_s(T)}{n} = 2\pi \int_{-\pi}^{\pi} d\Theta \frac{\Delta^2(\Theta) \cos^2 \Theta}{(\Delta^2(\Theta) + \tilde{\omega}_n^2)^{3/2}},$$

and

$$\beta(T) = 2\pi \Delta_0^2 \int_{-\pi}^{\pi} d\Theta \frac{\Delta^2(\Theta) \cos^4 \Theta}{(\Delta^2(\Theta) + \tilde{\omega}_n^2)^{3/2}}.$$

In order to model the varying crystal quality of the samples used to obtain the results shown in Fig. 1, we vary the impurity parameters $\Gamma$, $c$ and use Eq. (9)–(13) to extract the penetration depth $\lambda$ (at zero current) and the nonlinear current density scale $j_2$. In Fig. 2 we show $j_2/j_c$ at fixed temperature $T$ versus $\lambda(T)/\lambda(T = 0)$ obtained using this procedure. We show the results both for unitary scatterers and in the Born limit. We have superimposed the experimental data points from Fig. 1 by scaling them with $j_c = 150 \text{MA/cm}^2$ and $\lambda(T = 0) = 150 \text{nm}$ similar to the values given in the literature. As seen

Theory

The total current density can be written as

$$j = -2 j_c \int_{-\pi}^{\pi} \frac{d\Theta}{2\pi} \cos \Theta \int_{-\infty}^{\infty} \frac{d\omega}{\Delta_0} f(\omega) \left[ N_+(\Omega, \omega) - N_-(\Theta, \omega) \right],$$

where $f(\omega)$ is the Fermi function and $N_{\pm}(\Theta, \omega)$ denote the density of states for comoving and countermoving quasiparticles:

$$N_{\pm}(\Theta, \omega) = \text{Im} \frac{\tilde{\omega} \pm \Delta_0(j_s/j_c) \cos \Theta}{\sqrt{\Delta^2(\Theta) - [\tilde{\omega} \pm \Delta_0(j_s/j_c) \cos \Theta]^2}}.$$

Here, $j_s$ is the superfluid current density and $j_c$ the thermodynamic critical current density. In the following we will be interested in the current response of dirty superconductors and perform the calculations on the imaginary axis. Then the associated Matsubara frequencies $\tilde{\omega}_n$ are renormalized and must be determined from the following self-consistency conditions:

$$\tilde{\omega}_n = \omega_n + \Gamma \frac{g_0}{c^2 + g_0^2},$$

$$g_0 = \int_{-\pi}^{\pi} \frac{d\Theta}{2\pi} \sqrt{\Delta^2(\Theta) + \tilde{\omega}_n^2},$$

where $\omega_n = (2n + 1)\pi T$ denote the bare fermion Matsubara frequencies. As usual, $\Gamma = n_i/(\pi N_0)$ is the scattering rate proportional to the impurity concentration $n_i$, and $c = \cot \delta_0$, where $\delta_0$ is the s-wave scattering phase shift and $N_0$ is the density of states at the Fermi level. The strong scattering limit (unitary limit) is characterized by $c = 0$, whereas for the Born limit $c \gg 1$. For simplicity we have assumed a circular Fermi surface and study current flow along the nodal direction. The $d_{x^2-y^2}$-wave order parameter relevant for the high-$T_c$ cuprate materials can be written as

$$\Delta(\Theta) = \Delta_0 \sin(2\Theta).$$
The low (\(+\)) nonlinear current density scale shown in Fig. 1 and the theoretical results for the intrinsic nonlinear response in these materials, and allow one to extract information on the nature of the defects limiting the transport.

In summary, we have presented experimental results for the nonlinear current density scale \(j_2\) extracted from a range of different quality YBCO samples. We have argued that \(j_2\) is consistent with an intrinsic origin. A calculation including disorder within the self-consistent \(T\)-matrix approximation provides reasonable agreement with these experiments.

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In Fig. 2 the high temperature results agree reasonably well with the experiments. At high temperatures there is little difference between unitary and Born scatterers, as expected. For the dirtiest samples one can speculate whether effects other than the intrinsic nonlinearity become important. Certainly, the data point at \(\lambda = 480\,\text{nm}\) (see Fig. 1) seems to fall outside the fit, which may be related to the fact that this film was grown on sapphire.

The nonlinear current density scale \(j_2\) has a dramatically different dependence on \(\lambda\) at low temperatures \(T \ll T_{c0}\), where \(T_{c0}\) is the critical temperature of the clean system. Provided \(\max(n, T) \gg \Delta n_{s} / j_{2}\), where \(n = -\text{Im} \omega (\omega = 0)\) is the residual quasiparticle width, we can discuss this behavior within the quadratic approximation of Eq. (1). The functional form of the low \(T\) results shown in Fig. 2 can be understood in terms of Eq. (1) from Fig. 3 where we show the temperature dependence of the superfluid density \(n_s(T)\) and \(\beta(T)\) in the unitary limit for various \(\Gamma\). In the clean limit \(\Gamma = 0\), the \(\beta(T)\) coefficient diverges at low temperatures due to the nodes of the d-wave gap. With increasing disorder, this divergence is strongly suppressed. At higher temperatures, the \(\Gamma\) dependence of \(\beta(T)\) becomes much weaker. On the other hand, the superfluid density \(n_s\) in Fig. 3 exhibits a nonsingular dependence on \(\Gamma\). From Eq. (1) it is clear that the low \(T\) divergence of \(\beta(T)\) eventually will drive \(j_2\) down. This yields a prediction for low \(T \ll T_{c0}\) measurements valid if the samples with lowest penetration depth in Fig. 1 are clean enough to exhibit a pronounced \(\beta(T)\) upturn at low temperatures. It is clear from Fig. 2 that low temperature measurements will provide a more stringent test of the intrinsic nature of the nonlinear response in these materials, and allow one to

\[\text{FIG. 2: (Color online) Comparison of the experimental points shown in Fig. 1 and the theoretical results for the intrinsic nonlinear current density scale } j_2 \text{ versus the (zero-current) penetration depth } \lambda(T) / \lambda(T=0). \text{ The four curves correspond to unitary } (c=0) \text{ and Born } (c=4) \text{ scatterers in the low } (+, \times) \text{ and high } (\bigodot, \square) \text{ temperature limit.}\]

\[\text{FIG. 3: } n_s(T) \text{ and } \beta(T) \text{ versus temperature } T \text{ for a range of scattering strengths } \Gamma \text{ in the case } c=0. \text{ Here, } T \text{ and } \Gamma \text{ are given in units of the critical temperature of the clean system } T_{c0} \text{ and the critical scattering strength } \Gamma_{c0} \text{ at zero temperature, respectively. The right panel is similar to Fig. 6 of Dahm and Scalapino.}\]

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