Measurements of the Magnetic Field Dependence of $\lambda$ in YBa$_2$Cu$_3$O$_{6.95}$: Results as a Function of Temperature and Field Orientation

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(March 24, 2022)

We present measurements of the magnetic field dependence of the penetration depth $\lambda(H)$ for untwinned YBa$_2$Cu$_3$O$_{6.95}$ for temperatures from 1.2 to 70 K in dc fields up to 42 gauss and directions $0^\circ, \pm 45^\circ$ and $90^\circ$ with respect to the crystal b-axis. The experiment uses an ac susceptometer with fields applied parallel to the ab-plane of thin platelet samples. The resolution is about 0.15 Å in zero dc field, degrading to 0.2 or 0.3 Å at the higher fields. At low temperatures the field dependencies are essentially linear in H, ranging from 0.04 Å/gauss for $\Delta\lambda_0$ to 0.10 Å/gauss for $\Delta\lambda_6$, values comparable to the $T = 0$ Yip and Sauls prediction for a d-wave superconductor. However, the systematics versus temperature and orientation do not agree with the d-wave scenario probably due, in part, to residual sample problems.

74.25.Nf, 74.25.Ha, 74.72.Bk, 74.90.+n

When found, a successful theory for the unconventional superconductivity in the HiTc cuprates will have to incorporate a detailed knowledge of the structure of the pairing state. It is for this reason, that experiments studying the energy gap and its symmetry, and therefore the symmetry of the order parameter and pairing state, have always been of great importance. However, for sometime after the discovery of the HTSC’s, experiments probing the gap produced conflicting results and left much debate as to whether or not the pairing state had the conventional s-wave symmetry of BCS theory. Yip and Sauls [1] proposed experiments based on the nonlinear effects of an applied magnetic field on the Meissner state supercurrent that would differentiate between s-wave and d-wave superconductors (the d-wave state with $d_{x^2-y^2}$ symmetry having long been the favoured alternative to s-wave for the cuprates.) Today, however, this debate is largely settled despite the theory of the nonlinear Meissner effect remaining grossly under tested. Mounting evidence from several other experiments [2][4][4][4] has left little doubt that for HTSC’s the pairing state is essentially d-wave, exhibiting a predominantly $d_{x^2-y^2}$ symmetry.

Nevertheless, the work of Yip and Sauls and that of others [3][4][4] is still very much worth investigation. The nonlinear Meissner effect, as predicted, will lead to both a field dependent penetration depth as well as a magnetization that can contain a component transverse to the applied field. Furthermore, both quantities should show an anisotropy dependent upon field direction with respect to the nodes of a d-wave gap. In particular, for $\lambda(H)$ theory predicts that at sufficiently low temperatures there is a linear field dependence of the form

$$\lambda(T, H) - \lambda(T, 0) = \Delta\lambda(H) = \lambda(T)\alpha |H|/H_c(T),$$

where $\alpha$ is equal to unity for fields along a node and $1/\sqrt{2}$ when fields are along an antinode; the quantity $H_c(T)$ is a characteristic field of order the thermodynamic critical field $\lambda$. In contrast, a superconductor with a conventional gap would show an $H^2$ dependence of $\Delta\lambda$ with a thermally activated prefactor and with no anisotropy with respect to field direction. It is the prospect of using these measurements as a probe to determine the positions of the nodes that is now probably the most exciting aspect of this theory.

Unfortunately, experiment has lagged far behind the theory and node spectroscopy of this kind has never been successfully achieved. Experimenters face the difficult task of searching for a very small magnetic field effect hidden in a background of extrinsic field dependencies and other inherent sample anisotropies (for example, geometry, twinning, orthorombicity), all the while being restricted to the Meissner state, which limits the field range one can study. Early measurements of $\Delta\lambda(H)$ in YBa$_2$Cu$_3$O$_{7-\delta}$ [4][4][4] were limited to a resolution of $\sim 20$ Å (two orders of magnitude less than the method to be described in this paper), and neither investigated the anisotropy as a function of field in the ab-plane. The Minnesota group has concentrated on the angular dependence of the transverse magnetization $M_\perp$. Results for LuBa$_2$Cu$_3$O$_{7-\delta}$ did not support a pure d-wave pairing state, but measurements were dominated by the geometric demagnetization factor and trapped flux [2]. Subsequently, efforts have been made to alleviate the problems associated with sample geometry [3]. One cannot easily compare the sensitivity of the $M_\perp$ measurements to our $\Delta\lambda(H)$ measurements, but ultimately results from both types of experiment should complement each other. It is the aim of this paper to show that we have made substantial advances in testing the nonlinear Meissner effect, firstly by the development of a high resolution technique for measuring $\Delta\lambda(H)$ for fields in the ab-plane, and secondly by the demonstration that in YBa$_2$Cu$_3$O$_{6.95}$ the effect of magnetic fields on $\lambda$ is much smaller than that suggested by previous $\Delta\lambda(H)$ experiments.

Superconducting microwave cavity perturbation has proven to be a very successful technique for measuring small temperature dependencies in $\lambda$ of superconductors in the Meissner state [4][4]. However, this method is impractical for field dependent measurements. The application of a dc field inside the superconducting cavity...
is not possible, and the power dependence of the cavity severely inhibits studies as a function of microwave intensity. Here, an ac susceptometer has been designed that has very near the resolution of the microwave technique for variations in temperature ($\delta \lambda(T) \sim 0.15 \text{ Å}$), but can also be used to make field dependent measurements with only slightly less resolution ($\delta \lambda(H) \sim 0.2$ to 0.3 Å).

Such precise measurements of $\Delta \lambda(T, H)$ require the sample to be very well isolated from the background. Thermal isolation is achieved by mounting the sample on a sapphire cold finger in vacuum [7,8], a technique that is widely in use. Magnetic isolation is an extremely difficult condition to satisfy, since it is not possible to avoid exposing certain parts of the apparatus to the field as well; Figure 2 exhibits this problem quite clearly. The obvious approach is to use only 'nonmagnetic' materials in the construction of the apparatus, but at the level of sensitivity required here this did not sufficiently suppress a field dependent background signal. To overcome this problem, a custom made retractable sample holder was developed (see also Figure 3). Measurements of $\Delta \lambda(H)$ are made with a swept dc field; the amplitude of the ac field is fixed. The background signal is determined by extracting the sample from the susceptometer and repeating the same dc field sweep. Tests with only the sapphire sample plate and the equivalent amount of vacuum grease to hold a sample to the plate show no regular field dependence once the background is subtracted out. The peak values of the residual signal from this test correspond to a change of approximately 0.2 to 0.3 Å in penetration depth for a typical sized YBa$_2$Cu$_3$O$_{7-\delta}$ sample. We consider this to be our resolution in $\Delta \lambda(H)$.

The susceptometer operates with a 12 kHz ac field supplied by the primary coil. The output voltage from the secondary coils is proportional to the sample magnetization. To relate the change in voltage $\Delta v$ to a change in penetration depth $\Delta \lambda$ for a thin plate superconductor, we determine a calibration constant $k = \Delta \lambda / \Delta v$ valid in the limit $2 \lambda \ll t$, the sample thickness. (When $2 \lambda \sim t$ such a simple relation between $\Delta v$ and $\Delta \lambda$ does not exist.) This is done by heating the sample from the base temperature $T_b = 1.2$ or 4.2 K to above $T_c$; at 12 kHz $t << \delta$, the normal state skin depth, so the resulting change in voltage going from the superconducting to the normal state $v_{sn}$ can be related to the change in the effective superconducting volume of the sample $\approx V - 2 \lambda(T_b)$, where $A$ is the sample area and $V$ its volume both at $T_b$.

The calibration constant $k$ is then

$$k = (t - 2\lambda(T_b)) / 2v_{sn} \approx t / 2v_{sn}$$

Measurements were made on a detwinned single crystal of YBa$_2$Cu$_3$O$_{6.95}$ grown in an yttria-stabilized crucible [9]. The sample had dimensions 0.98 : 1.89 : 0.056 in mm. It had a $T_c = 91.8$ K, also measured using the ac susceptometer, and exhibited the characteristic linear $\lambda(T)$ behavior of a high quality cuprate superconductor at low temperatures. Figure 4 shows a graph of $\Delta \lambda_a$ and $\Delta \lambda_b$ as a function of temperature. For both quantities the linear $\Delta \lambda(T)$ crosses over to higher powers in $T$ below 4.5 K. This crossover is due to an impurity level that is typical of crystals grown in yttria-stabilized crucibles [2] and is not believed to be large enough to mask the nonlinear Meissner effect [3].

The crystal was initially mounted so the fields were parallel to the $b$-axis in the $ab$-plane. It was subsequently rotated in three 45° increments around the $c$-axis. An extensive set of data was taken for each crystal orientation. Measurements of $\Delta \lambda(T)$ were made in zero dc field from 1.2 K to 20 K; the ac drive field had a peak amplitude of $\approx 7$ gauss. Measurements of $\Delta \lambda(H)$ were made for a series of fixed temperatures from 1.2 K to 70 K with the same amplitude for the drive field. The dc field was stepped in even increments through a loop of $\pm 42$ gauss. Data was collected for 10 successive loops and averaged. Similar runs were done with the sample extracted from the susceptometer in order to determine the background. A background run was not taken after every data run and often the same background was used for several data sets. However, experimental conditions were never allowed to change markedly before a background run was performed, and careful attention to the signal gain of the system showed that this time saving measure did not introduce any noticeable error into our analysis.

As mentioned, the results for $\Delta \lambda_a(T)$ and $\Delta \lambda_b(T)$ are shown in Figure 2. Results for the ac field 45° to the crystal axes (data not shown) also exhibit the linear $T$ dependence along with the crossover to higher powers below about 4.5 K. Linear fits to $\Delta \lambda(T)$ above 4.5 K give slopes of 5.0 Å/K and 4.6 Å/K for the $a$- and $b$-directions respectively, and 4.7 to 4.8 Å/K for the 45° orientations. The magnitudes of these slopes are typical for YBa$_2$Cu$_3$O$_{6.95}$, as is the result that $\Delta \lambda_a(T) / \Delta T > \Delta \lambda_b(T) / \Delta T$. Furthermore, as expected, the values for the 45° orientations are averages of the values for the $a$- and $b$-directions. These results give us confidence that the zero field electrodynamics of our crystal are no different from previously published data and furthermore that we have not misidentified the $a$- and $b$-axes.

The results for $\Delta \lambda(H)$ are shown for the $a$- and $b$-directions in Figure 3. The results for $\pm 45^\circ$, shown in Figure 4, should be identical for a rectangular shaped orthorhombic crystal; clearly they are not. In each graph the data sets have been separated by 1 Å at zero field for the purpose of clarity. The 1.2 K data has not been plotted, as it is not distinguishable (within resolution) from the 4.2 K results. All other data from 4.2 K to 70 K is presented, and it is clear from this that the effect of field on $\Delta \lambda$ increases with temperature. This temperature dependence is clearly not exponential, which is consistent with the belief that the energy gap in YBa$_2$Cu$_3$O$_{6.95}$ is not s-wave. The strongest test of a d-wave gap within the theory of the nonlinear Meissner effect is the linear field dependence in $\Delta \lambda$. This is seen for $\Delta \lambda_a$ over most of the temperature range studied. For the other sample orientations, it is only at low temperatures where the field de-
dependence is essentially linear. Linear fits to the 4.2 and 10 K data give slopes of about 0.04 and 0.10 A/gauss for the a- and b-directions, respectively, and about 0.07 A/gauss for the ±45° directions. In contrast, measurements at 12 K by Maeda et al. [1] (measured for fields perpendicular to the ab-plane) gave a linear field dependence of ~8 A/gauss, roughly 100 times larger than our result. In even greater contrast is the quadratic field dependence seen by Sridhar et al. [10] in 1989, but this is most likely attributable to the general unavailability of high quality crystals at that time.

Although this experiment has improved the sensitivity of Δλ(H) measurements by nearly two orders of magnitude and shown that the nonlinear effects are much smaller than previously reported, it is clear that we have not yet reached the intrinsic values, at least in the ±45° and b-directions. First of all, the fact that the +45° and −45° measurements do not agree shows us that the results do not follow the symmetry of a rectangular shaped, orthorhombic crystal. Possible mechanisms for this result are remnant microtwins from the detwinning process, or sharp-edge effects that are not symmetrically distributed. The much larger slope for the b-direction is also surprising, and possibly due to some nonlinear effect associated with the Cu-O chains. However the a-axis results, being the most linear in H and having the weakest temperature effects, are likely closest to the intrinsic values.

Xu, Yip and Sauls (XYS) [5] define the characteristic field H_o = (3v_o/2c) tan(2λ), where v_o ∼ Δ_o/v_f, the gap maximum divided by the Fermi velocity; they estimate H_o to be approximately 2.5 T. To extract H_o from our a-direction data we rearrange Equation 1 as H_o = λ_o/αexp(2λ/Δ_o), where the quantity in brackets is the reciprocal of the slope of the plot to find the characteristic field of 2.8 T for temperatures in the range 1.2 to 10 K. However, as discussed in XYS [6], thermal effects cause the T=0 linear field dependence to crossover to quadratic; the crossover field being given by H_T = (T/Δ_o)H_o equals 60 gauss at our lowest temperature of 1.2 K. Thus the full Yip and Sauls theory predicts a weaker, quadratic dependence of Δλ(H) in our temperature range, and the above agreement is probably accidental.

In summary, we have presented measurements of Δλ(H) for an untwinned crystal of YBa_2Cu_3O_6.95 made as a function of temperature and, for the first time, as a function of field orientation in the ab-plane. The resolution of this experiment is ~100 times greater than any previous experiment measuring Δλ(H). At low temperatures a linear field dependence in Δλ(H) is observed, which while apparently agreeing with the T=0 limit of the Yip and Sauls theory is not in agreement when thermal effects are taken into account. Also, there is an anisotropy in Δλ(H) for fields ±45° to the b-axis that can only be explained by some mechanism that breaks the crystal symmetry; we suggest remnant twinning, or unsymmetrical edge effects.

ACKNOWLEDGMENTS

The authors acknowledge the assistance of S. Kamal and P. Dosanjh. This work was supported by the National Science and Engineering Research Council of Canada and the Canadian Institute for Advanced Research. DAB gratefully acknowledges support from the Sloan Foundation.

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FIG. 1. The ac susceptometer: constructed from only materials with very small magnetic susceptibilities. The sample is shown extracted from the pick-up coil; the dashed outline shows the sample in the measurement position.

FIG. 2. $\Delta \lambda(T)$ and the superfluid fraction $\lambda^2(0)/\lambda^2(T)$ in zero dc field for a-(circles) and b-(squares) directions.

FIG. 3. $\Delta \lambda$ as a function of $H$ for fields parallel (top) and perpendicular (bottom) to the crystal b-axis.
FIG. 4. $\Delta \lambda$ as a function of $H$ for fields $\pm 45^{\circ}$ to the crystal axes.