Magnetic Field Dependence of λ in $YBa_2Cu_3O_{6.95}$: Results as a Function of Temperature and Field Orientation

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We present measurements of the magnetic field dependence of the penetration depth $\lambda(H)$ for an untwinned $YBa_2Cu_3O_{6.95}$ single crystal for temperatures from 1.2 to 7 K in dc fields up to 177 G and directions 0°, ±45°, and 90° with respect to the crystal b axis. The edges of the platelet sample have been polished round to limit extrinsic effects. At 1.2 K, $\Delta\lambda(H)$ in the a and b directions could be interpreted as agreeing with the Yip and Sauls prediction for the nonlinear Meissner effect in a d-wave superconductor. However, the systematics versus temperature and orientation, a key aspect of the theory, do not agree, and we conclude that the nonlinear Meissner effect is suppressed by a factor of 10 or larger.

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In high-T_c superconductors, experiments studying the energy gap and its symmetry have always been of great importance. However, early experiments 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 state with $d_{x^2-y^2}$ symmetry having been the favored alternative to s wave for the cuprates.) This debate has now been largely settled by other experiments [2–4], which have left little doubt that for HTSC's the pairing state exhibits a predominantly $d_{x^2-y^2}$ symmetry. However, the nonlinear Meissner effect (NLME) remains experimentally undertested, although there has been a considerable amount of theoretical work [5–10].

The NLME leads to a field dependent penetration depth and magnetization, and these effects depend on the orientation of the field relative to the $d_{x^2-y^2}$ nodes. In particular for $\lambda(H)$, theory predicts that at low temperatures there is a linear field dependence of the form

$$\lambda(T,H) - \lambda(T,0) = \Delta\lambda(H) = \lambda(T)\alpha|H|/H_o\,, \quad (1)$$

where α is equal to unity for fields along a node and $1/\sqrt{2}$ when fields are along an antinode; the quantity H_o is a characteristic field of the order of the thermodynamic critical field [5]. 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. Using these measurements to determine the positions of the nodes is one reason researchers have remained interested in this theory.

Unfortunately, *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, orthorhombicity), all the while being restricted to the low field range of the Meissner state. Early measurements of $\Delta\lambda(H)$ in $YBa_2Cu_3O_{7-\delta}$ [11,12] were limited to a resolution of ~20 Å and the effects that were seen are now known to be of some extrinsic origin. This was demonstrated recently by two independent research teams, ourselves [13] and Carrington *et al.* [14]. Both groups have developed high precision techniques capable of measuring $\Delta\lambda(H)$ to within a few tenths of an angstrom, and both measured the effect in $YBa_2Cu_3O_{6.95}$ to be much less than the resolution of the previous experiments. While neither of these recent measurements could claim to have seen the intrinsic field dependence of λ in $YBa_2Cu_3O_{6.95}$, they have clearly identified a more appropriate scale on which this search must take place. A complementary test of the theory has been carried out by the Minnesota group on the angular dependence of the transverse magnetization $M_\perp$. Early results for $LuBa_2Cu_3O_{7-\delta}$ did not support a pure d-wave pairing state, but measurements were dominated by the geometric demagnetization factor and trapped flux [15]. Subsequently, efforts were made to alleviate the problems associated with sample geometry [16], and improved measurements of $M_\perp$ for a disk shaped single crystal of $YBa_2Cu_3O_{6.95}$ have recently been reported [17]. Within their experimental uncertainty, they did not observe the fourfold symmetry in $M_\perp$ expected for the NLME in a $d_{x^2-y^2}$ superconductor. While one cannot easily compare the sensitivity of the $M_\perp$ measurements to our $\Delta\lambda(H)$ measurements, results from both types of experiment should ultimately complement each other under a single consistent explanation. It is the aim of this paper to show that we have made substantial advances in testing the nonlinear Meissner effect, demonstrating that $\Delta\lambda(H)$ does not have the dependence on temperature or field direction predicted by Yip and Sauls.

We employ a custom susceptometer, which is of standard design but with the counterwound pickup coils mounted on a precision ground high purity sapphire form; this serves both to reduce the magnetic field dependent background and to suppress thermal coupling between the sample and the susceptometer to negligible values. The high sensitivity was achieved through a combination of a relatively good filling factor, a liquid helium cooled transformer, and low noise external compensation and amplification. The small residual field dependent background is determined *in situ* using a retractable sample holder. The sample is mounted on a thin sapphire plate whose temperature can be accurately set [18].

The susceptometer operates with a 12 kHz ac field supplied by the drive coil. To relate a change in voltage Δv from the pickup coils to a change in penetration depth $\Delta\lambda$ for a thin plate superconductor, we determine a calibration constant $k = \Delta\lambda/\Delta v$ that is valid only in the limit $2\lambda \ll t$, the sample thickness. 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 $t \ll \delta$, the normal state skin depth, so the resulting change in voltage v_{sn} (superconducting to normal) can be related to the change in the effective superconducting volume of the sample $\simeq V - 2A\lambda(T_b)$, where A is the sample area and V its volume both at T_b. The calibration constant k is

$$k = [t - 2\lambda(T_b)]/2v_{\mathrm{sn}} \simeq t/2v_{\mathrm{sn}}\,, \qquad (2)$$

and is determined for each run.

Measurements as a function of field are done by stepping the dc magnet current in small increments through many successive cycles from positive to negative fields, with the ac field amplitude fixed. Data are collected and averaged for each field increment over several cycles. To determine the background signal (which is hysteretic), similar runs are done with the sample extracted from the susceptometer. The corrected signal (background subtracted out) for a sample in the Meissner state does not show any measurable hysteresis above our resolution.

Measurements were made on a detwinned single crystal of $YBa_2Cu_3O_{6.95}$ grown in an yttria-stabilized zirconia crucible [19]. The platelet sample had dimensions a:b:c of 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 linear $\Delta\lambda(T)$ of a high quality cuprate superconductor at low temperatures. The inset of Fig. 1 shows the superfluid fraction $\lambda^2(0)/\lambda^2(T)$ for the a and b directions. Both quantities deviate from linearity below 4 K, which is due to an impurity level typical of crystals grown in yttria-stabilized zirconia crucibles [20] and is not believed to be large enough to mask the nonlinear Meissner effect [5]. Measurements of $\Delta\lambda(T)$ were also made along directions $\pm45°$ to the b axis, and as expected were the average of $\Delta\lambda_a(T)$ and $\Delta\lambda_b(T)$. These results lend confidence that the zero field $\lambda(T)$ of this crystal is not different from previously published data.

After this characterization, the crystal's sharp edges were removed by abrasion on a 0.1 micron grit diamond polishing pad. The rectangular shape of the crystal was not altered, the ab faces were not allowed to touch the pad, and observation under a microscope showed the polished edges to be optically smooth. All of the $\Delta\lambda(T)$ measurements were then repeated. The a direction results in Fig. 1 clearly show the preservation of the low temperature behavior, with no sign of the sample crossing over to a *dirty d-wave* superconductor [21], which indicates that it was not damaged. Similar results were seen for the other directions.

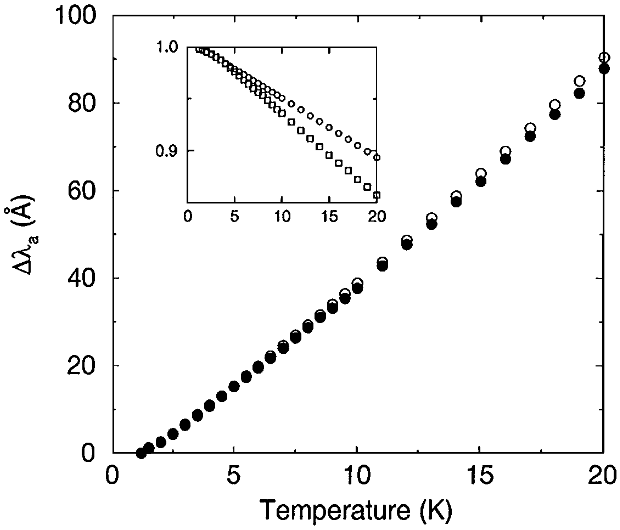

FIG. 1. $\Delta\lambda_a$ as a function of T for sample as-grown (open circles) and with polished edges (filled circles). The inset shows the superfluid fraction $\lambda^2(0)/\lambda^2(T)$ for the as-grown sample in the a (circles) and b (squares) directions.

Polishing the edges of the crystal does have a dramatic effect on the nonlinear response. Figure 2 shows measurements of $\Delta\lambda(H)$ for fields $\pm45°$ to the b axis in the ab plane with the sample at 60 K. Results in the as-grown sample clearly break the symmetry of the situation and can be attributable only to some extrinsic effect. After polishing, $\Delta\lambda(H)$ agrees for both directions, as it must for a rectangular orthorhombic crystal. Also note the order of magnitude reduction in the signal, revealing the ±0.1 Å resolution achieved in our measurements. Similar improvements were seen at lower temperatures as well, and with no changes upon further polishing of the edges.

Using the crystal with polished edges, measurements were made to test the NLME. The crystal was initially mounted with the fields parallel to the b axis. It was subsequently rotated in three 45° increments around the c axis; the orientation of the sample was accurate to within a few degrees. An independent calibration was done for each orientation, and measurements of $\Delta\lambda(H)$ were made at a fixed temperature of 1.2 K. The dc field was stepped in even increments through a loop of ±177 G; the ac drive field had a peak amplitude of 2.5 G. All measurements are done below $H_{c1}(T)$ [22]; tests using maximum fields of 42, 93, 140 G, each with the sample first cycled above T_c, gave identical results.

The results for $\Delta\lambda(H)$ as a function of orientation are shown in Fig. 3. The top panel shows the results for the

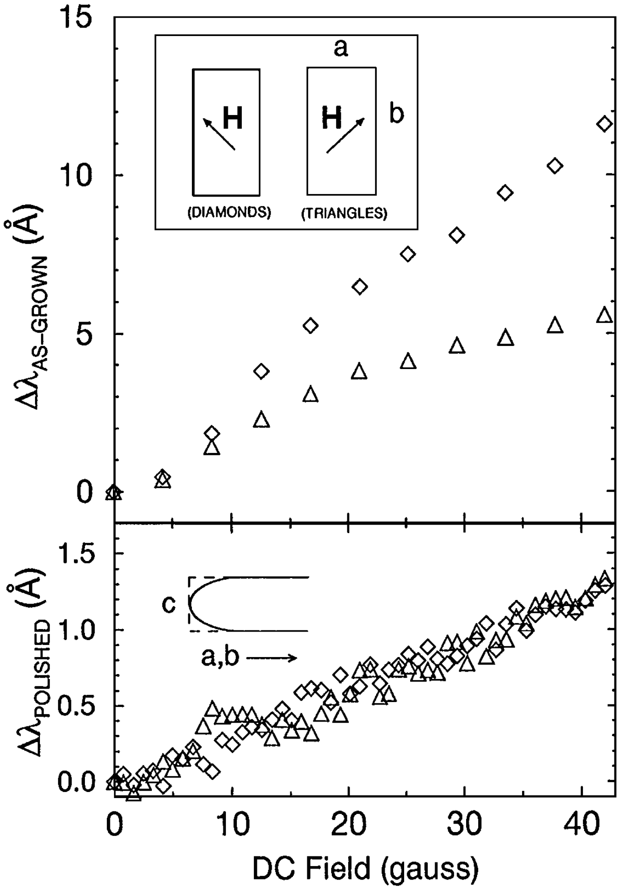

FIG. 2. $\Delta\lambda(H)$ at 60 K for H applied $\pm 45°$ to the crystal b axis (see inset). Top panel: results for the as-grown crystal. Bottom panel: results after polishing the edges of the crystal. Also shown is the shape of an edge before and after polishing.

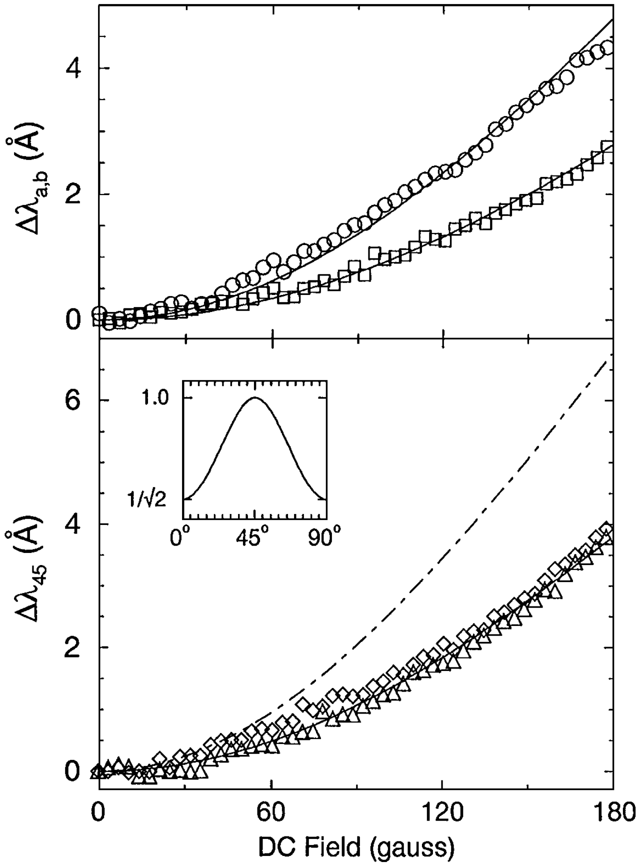

FIG. 3. $\Delta\lambda$ as a function of $\vec{H}$ at 1.2 K. Top panel: $\Delta\lambda_a$ (circles) and $\Delta\lambda_b$ (squares) with theoretical curves for H along antinodes. Bottom panel: $\Delta\lambda_{\pm 45}$; the dashed line is the theoretical curve for H along a node, whereas the solid line is the average of the two curves in the top panel. The inset shows how α varies with field orientation with respect to an antinode.

a and b directions; $\Delta\lambda_a(H)$ is larger than $\Delta\lambda_b(H)$ as one would expect since λ_o is larger in the a direction. The bottom panel shows the results when the field is applied $\pm 45°$ to the b axis, and as should be the case, the two sets of data agree to within error. The inset in the bottom panel gives the angular dependence of the prefactor α in Eq. (1) [23] and is there to show that small uncertainty in our sample orientation would not introduce any significant error.

Included with our data in Fig. 3 are theoretical curves for the NLME at 1.2 K generated by the numerical solution given by Li *et al.* [9]. Parameters needed to produce these curves are the zero temperature penetration depth λ_o, an energy scale Δ_o related to the gap maximum, and the characteristic field H_o [24]. Using literature values of 1600 and 1030 Å for λ_o in the a and b directions [25], the values of Δ_o are determined for this crystal by matching the slope of its $\Delta\lambda(T)$ data with the Li result for $\Delta\lambda$ in zero field. This gave a Δ_o of 220 and 155 K for the a and b direction, respectively, which seem reasonable for YBCO [5]. Values of 2.03 and 1.85 T for H_o gave the closest agreement with the $\Delta\lambda_a(H)$ and $\Delta\lambda_b(H)$ data, respectively, in the top panel of Fig. 3. These are close to Yip and Sauls' estimate of 2.5 T, which initially suggests that we are seeing the expected NLME [5]. However, the bottom panel of Fig. 3 shows that $\Delta\lambda_{45}(H)$ is simply the average of the the values observed in the a and b directions (solid line), and does not show the enhancement expected in the node direction (dashed line), which is the main tenet of the Yip-Sauls theory. We emphasize that this result holds for all fields and temperatures where measurements were taken, and therefore the missing enhancement is unlikely to be due to an extrinsic effect. Based on the scatter in the data, this result translates into an order of magnitude reduction in the enhancement effect. To the best of our knowledge this is the first experiment to directly test this aspect of the Yip-Sauls theory [26].

Figure 4 shows the evolution of $\Delta\lambda_a(H)$ with T. For clarity the 1.2 K data is not replotted, only the best fitting theoretical curve. Theoretical curves at 4.2 and 7.0 K show a strong suppression of $\Delta\lambda(H)$ with increasing T, in stark contrast to the measurements, which show a small

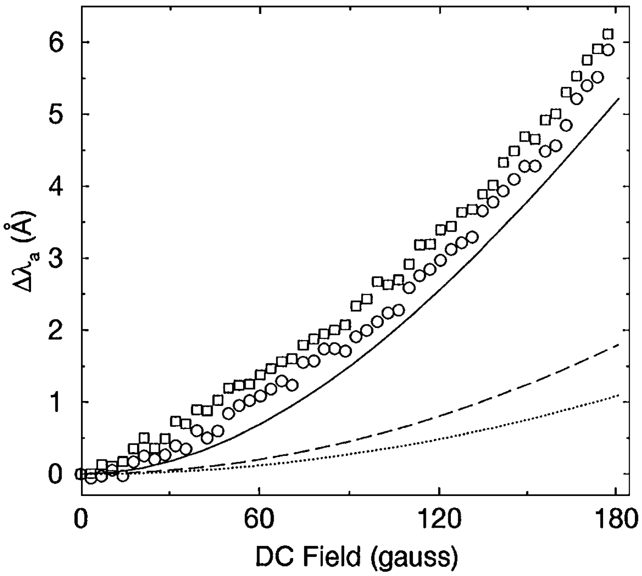

FIG. 4. $\Delta\lambda_a$ as a function of H at various temperatures. Data: 4.2 K (circles), 7.0 K (squares). Theoretical curves: 1.2 K (solid line), 4.2 K (dashed line), 7.0 K (dotted line).

increase. This behavior has been seen before [13,14] at temperatures of 10 K and above. However, measurements at lower temperatures using a crystal with polished edges to control extrinsic effects have provided convincing evidence that for $YBa_2Cu_3O_{6.95}$ the Yip-Sauls NLME is not the source of the observed $\Delta\lambda(H)$ above 1.2 K.

In summary, we have presented measurements of $\Delta\lambda(H)$ for an untwinned crystal of $YBa_2Cu_3O_{6.95}$ made as a function of temperature and of field orientation in the *ab* plane. We have shown that the sharp edges of an as-grown crystal may introduce extrinsic effects even in geometries where on average the demagnetizing effects are small. A careful comparison of our results has been made with theory, and while we acknowledge that a more complete theory should also include the orthorhombicity of the crystal, the overwhelming disagreement we see renders a more complicated analysis unnecessary at this time. In particular, while at 1.2 K we can describe our a- and b-axis data quite well with the Yip-Sauls theory using reasonable values for the characteristic field H_o, for the field applied 45° to these directions an enhancement in $\Delta\lambda(H)$ is not observable above our resolution, which implies that it is at least an order of magnitude less than predicted. (This is consistent with an *upper bound* of 30%, the predicted $M_\perp$ imposed by "noise" in the Minnesota group's latest null result [17].) We have also observed that $\Delta\lambda(H)$ increases slightly with temperature just above 1.2 K, whereas theory predicts a strong suppression. It appears, therefore, that if present, the nonlinear Meissner effect of Yip and Sauls has been suppressed by a factor of the order of 10 or larger.

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[23] The angular dependence of α is given in [9]. Note: α may differ from the value given in [5] depending on the definition of λ and the measurement technique. In Ref. [5], λ is defined by the initial rate of decay of the magnetic field inside the sample. For a volume exclusion experiment, such as ours, a more appropriate definition is $\lambda = \int_0^\infty H(z)/H(0)\,dz$, which reduces $\Delta\lambda(H)$ by a factor of 2. On the other hand, an ac measurement results in a $\Delta\lambda(H)$ twice that of a dc measurement. Thus, for our ac susceptibility, the two factors cancel and the appropriate α is numerically equal to the value in [5].
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