# Anisotropic Nature of High-Temperature Superconductivity in Single-Crystal $Y_1Ba_2Cu_3O_{7-x}$

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We report the first contact-free measurements of the upper critical fields, $H_{c2}(T)$, of single-crystal $Y_1Ba_2Cu_3O_{7-x}$. In contrast to reported resistive measurements, we find that the anisotropy near $T_c$ is temperature independent in agreement with anisotropic Ginzburg-Landau theory. Estimates of the anisotropic Ginzburg-Landau and London parameters are reported. These indicate that despite the large anisotropy in $H_{c2}$ (5:1), the inferred low-temperature interplanar coherence length ($\xi_z = 7$ Å) remains larger than the Cu-O layer spacing of 3.9 Å. Superconductivity in $Y_1Ba_2Cu_3O_{7-x}$ thus remains fundamentally three dimensional in nature for a substantial temperature range below $T_c$.

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Despite the flurry of activity on high-$T_c$ superconductivity in Cu-O based perovskite-type materials that has followed the breakthrough discovery of superconductivity at $\simeq 35$ K in $La_{2-x}Ba_xCuO_4$ by Bednorz and Müller,[1] the macroscopic nature of the superconductivity in these new materials and the mechanism responsible for their superconductivity are not yet clear. The availability of single crystals of the new Cu-O based superconductors, and the anisotropy manifest in their resistivity,[2] critical current density,[3] and upper[2,4-6] and lower[3] critical fields, establish the importance of anisotropy to the macroscopic nature of the high-temperature superconductivity. In this paper we report the first inductive measurements of $H_{c2}$ as a function of orientation for single-crystal $Y_1Ba_2Cu_3O_{7-x}$. We use this and other data to estimate the anisotropic Ginzburg-Landau parameters, and to examine the degree to which existing theories of anisotropic superconductors are able to describe the observed behavior of $Y_1Ba_2Cu_3O_{7-x}$. We find that the anisotropic Ginzburg-Landau theory describes our data well near $T_c$, and that although the superconductivity in $Y_1Ba_2Cu_3O_{7-x}$ is strongly anisotropic, it is three dimensional in nature. Elaborations of available theories are needed to explain lower-temperature features of our data.

The $Y_1Ba_2Cu_3O_{7-x}$ single crystal used in this study was obtained from a partially melted pellet made with an off-stoichiometric composition according to a procedure described by Dinger *et al.*[3] This procedure routinely resulted in highly faceted crystals with dimensions of approximately 200 $\mu$m, with occasional crystals approaching 0.5 mm in size. The crystal studied here had dimensions of 340 by 280 by 160 $\mu$m.[3] Measurements in a scanning x-ray diffractometer showed this crystal to be of high quality with unit-cell dimensions of $a = 3.83$ Å, $b = 3.89$ Å, and $c = 11.71$ Å. The degree of orthorhombicity was $b/a = 1.016$. The crystal was macroscopically twinned in the $a$-$b$ plane. The diffraction peak widths were limited by instrumental resolution, indicating domains in excess of 500 Å in size and little compositional variation.

The inductive transition of the crystal was measured by means of a variation of a technique used by Dalrymple and Prober.[7] The crystal was tightly wrapped with eight turns of 30-$\mu$m-diam insulated copper wire. A 200-pF capacitor was connected in parallel with this inductor to form an $LC$ circuit which was resonant at about 70 MHz. The wrapped crystal, capacitor, and a carbon-glass thermometer (Lake Shore Cryotronics, encapsulation removed) were all affixed to an alumina TO-5 header which was mounted in a rotating fixture insert of a 15-T superconducting magnet. Variable temperatures were provided by helium gas flowing through a heated copper block below the sample. A Minicircuits ZSC-2-1 signal splitter, used as a directional coupler, and a HP4194A gain-phase analyzer were used to measure the reflected signal from the resonant circuit, as shown in the inset to Fig. 1(a). As the sample was slowly cooled through the transition, the change in inductance resulted in a change in resonant frequency. The transition temperature was determined by the measurement of the reflected power as a function of temperature at a fixed frequency. The frequency was chosen at the steepest point on the amplitude-versus-frequency curve above the transition temperature. The field due to the measurement current is estimated to be $\simeq 3$ G; smaller current values only reduced the signal-to-noise ratio. Figures 1(a) and 1(b) show a selection of the curves of the reflected power versus temperature for both parallel and perpendicular orientation. Data were taken for both warming and cooling to verify that the sample and the thermometer were at the same temperature. Each curve represents $\simeq 2-3$ h of data taking depending on the temperature span required.

The transition temperature at a given field was determined by the construction shown in the figures. Although our technique for extracting $T_c$ is somewhat arbitrary, the transitions are well defined and broaden only slightly with increasing field; any reasonable technique will yield same values for $dH_{c2}/dT$. The temperatures were corrected for magnetoresistance with use of data from Sample, Brant, and Rubin,[8] the corrections

 

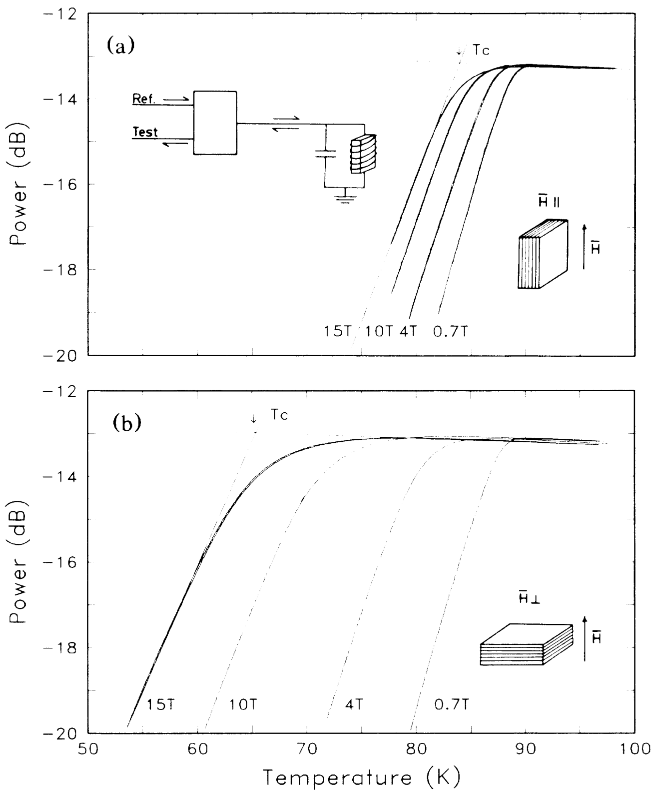


FIG. 1. Temperature dependence of the amplitude of the reflected signal from a resonant tank circuit with the $Y_1Ba_2Cu_3O_{7-x}$ crystal mounted with the Cu-O planes (a) parallel and (b) perpendicular to the applied field, for $H=0.7$, 4, 10, and 15 T. The constructions indicate how $T_c$ at a given field was determined. Inset in (a): Schematic of the measurement apparatus.

amounting to $\simeq 1.5$ K at the highest field.

Figure 2 shows the temperature dependence of $H_{c2}$ for the Cu-O planes oriented parallel and perpendicular to the applied field. The zero-field transition temperature extrapolated from both the parallel and the perpendicular data is 88.8 K. The temperature dependence of $H_{c2}^{\parallel}$ is basically consistent with a straight line with a slope of $-2.3$ T/K, with the highest-field points possibly indicating some upward curvature. Extrapolation of the $H_{c2}^{\parallel}$ curve back to zero temperature according to the dirty-limit isotropic formula (with no Pauli paramagnetism limiting effects),[9] $H_c(0)=0.69T_c\,dH_c/dT$, gives $H_{c2}^{\parallel}(0)=140$ T. {For a strongly anisotropic superconductor a more appropriate extrapolation might be linear [which would give $H_{c2}^{\parallel}(0)=204$ T], and the correct extrapolation might be even higher than linear.} In contrast, the temperature dependence of the perpendicular field $H_{c2}^{\perp}$ shows a pronounced upward deviation from a linear dependence. Near $T_c$, $H_{c2}^{\perp}$ has a slope of $-0.46$ T/K, but below 78 K the data are consistent with a slope of $-0.71$ T/K. Extrapolating these two curves back to zero temperature using the isotropic dirty-limit formula,

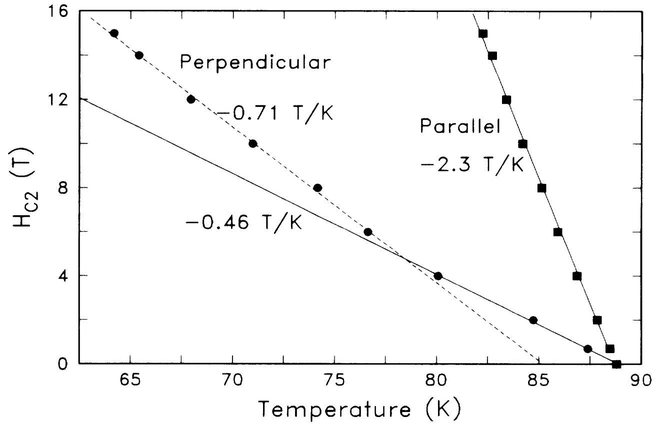


FIG. 2. Temperature dependence of the $H_{c2}$ for the $Y_1Ba_2Cu_3O_{7-x}$ crystal oriented with the Cu-O planes parallel and perpendicular to the applied field.

we get estimates for $H_{c2}^{\perp}(0)$ of 29 and 42 T.

Figure 3 shows the angular dependence of $dH_{c2}/dT$ for an applied field of 10 T. Here we define $dH_{c2}/dT$ as 10 T divided by the difference of the transition temperature at 10 T at a given angle and the zero-field transition temperature. The sharpness of the angular data near parallel orientation underscores the need for precise alignment of the sample. The two curves plotted in the figure are the angular dependences predicted for $H_{c2}$ at fixed temperature by the anisotropic Ginzburg-Landau theory,[10] $H_{c2}(\theta)=H_{c2}^{\perp}[\sin^2\theta+(m_\perp/m_\parallel)\cos^2\theta]^{-1/2}$, for two values of $m_\perp/m_\parallel$. The value which fits the end points results in a curve that does not fit the data near the peak, and a value which fits the data near the peak results in a curve that is significantly below the data at

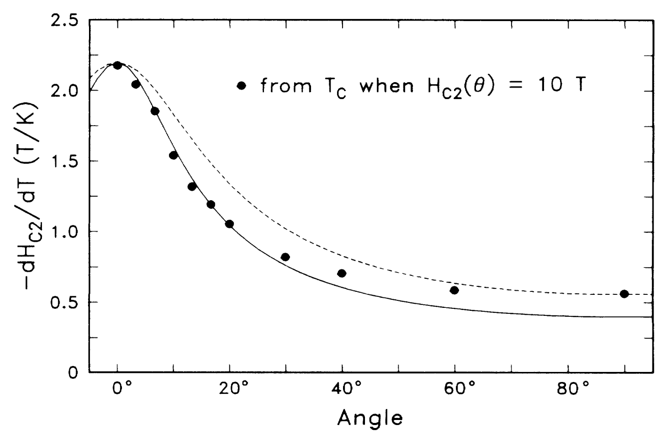


FIG. 3. Dependence of $dH_{c2}/dT$ on angle between the Cu-O planes and the applied field as calculated from data taken at 10 T. The two curves are the angular dependences predicted by the anisotropic Ginzburg-Landau theory for two sets of anisotropy parameters.

large angles. The deviation of the data from the latter theoretical curve would be significantly less if there were no break in the slope of $H_{c2}^{\perp}$ at $\simeq 78$ K.

We looked for, but did not observe, any change in the transition temperature for rotations of 0°, 45°, and 90° in the $a$-$b$ plane in a field of 10 T. We believe that we would have seen an anisotropy in our crystal if it were as large as the $\simeq 1$-K change reported by Hidaka *et al.*[5] for their resistive measurements on single-crystal $Y_1Ba_2Cu_3O_{7-x}$ with a 70-K transition temperature. Iye *et al.*[6] likewise did not observe any $a$-$b$ anisotropy in their resistive measurements on a $\simeq 90$-K $Y_1Ba_2Cu_3O_{7-x}$ crystal.

Our results do not show any of the curvature near $T_c$ that was evident in the resistive measurements of $dH_{c2}/dt$ by Hidaka *et al.*[2] and Iye *et al.*[6] on single-crystal $Y_1Ba_2Cu_3O_{7-x}$ and in previous resistive measurements on $La_{2-x}Ba_xCuO_4$ by Shamoto, Onoda, and Sato[4] and Hidaka *et al.*[5] The resistive measurements are made by passing currents along the Cu-O planes and necessarily involve currents flowing on many planes. We know from our earlier work[3] that the critical currents at high temperature and fields between the planes are very small. These low critical currents and the possibility of damaged surface layers could be complicating the determination of $H_{c2}$ from resistive measurements.

Our measured values for some of the anisotropic parameters of $Y_1Ba_2Cu_3O_{7-x}$ are given in Table I along with various derived quantities. The coherence length in the Cu-O plane, $\xi_0$, is calculated from the estimate of $H_{c2}^{\perp}(0)=29$ T and the relation[10] $H_{c2}^{\perp}(0)=\phi_0/2\pi\xi_0^2$. The ratio of $H_{c2}^{\parallel}/H_{c2}^{\perp}=\xi_z/\xi_0$ results in $\xi_z=7$ Å. The coherence length perpendicular to the Cu-O layers, $\xi_z$, is significantly greater than the spacing between the Cu-O layers, 3.9 Å, indicating that although the superconductivity is strongly anisotropic, it remains three dimensional in nature down to very low temperatures. According to the Josephson-coupled layer model, the crossover to two-dimensional behavior is expected when $\xi_z=s/\sqrt{2}=2.8$ Å, where $s$ is the Cu-O interplanar spacing.[11] The measurements of Freitas, Tsuei, and Plaskett[12] showed that the fluctuations near $T_c$ were also three dimensional in nature. $H_{c1}^{\perp}$ was used to calculate the penetration depth in the Cu-O planes, $\lambda_0$, from the relation $H_{c1}^{\parallel}=(\phi_0/4\pi\lambda_0^2)\ln(\lambda_0/\xi_0)$. From the penetration depth and $H_{c2}^{\perp}$ we calculate $H_c(0)=H_{c2}^{\perp}/\sqrt{2}\kappa_0$, where $\kappa_0=\lambda_0/\xi_0$. The values for $\kappa_z$ and $\lambda_z$ are calculated from[13] $\kappa_z=(m_{\parallel}/m_{\perp})\kappa_0$ and $\kappa_z=(\lambda_z\lambda_0/\xi_z\xi_0)^{1/2}$. For these calculations we have used the anisotropy ratio given by the temperature dependence of $H_{c2}$ near $T_c$ and the dirty-limit isotropic relationship between the high-temperature data and the low-temperature parameters. Obviously, use of the larger anisotropy observed for $H_{c1}$ would alter the numerical results as would the use of extrapolations other than the conventional isotropic dirty-limit relationship.

TABLE I. Measured and derived anisotropic parameters for single-crystal $Y_1Ba_2Cu_3O_{7-x}$. Parallel and perpendicular refer to the direction of the field applied relative to the copper-oxygen planes. For the case of $J_c$, the currents are actually flowing perpendicular to the applied field.

| | Parallel | Perpendicular |
|---|---|---|
| Measured parameters | | |
| $T_c$ | 88.8 K | |
| $(dH_{c2}/dT)(T_c)$ | 2.3 T/K | 0.46 T/K |
| $H_{c1}$(4.5K) | $\leq 0.005$ T | 0.5 T |
| $J_c$(0 T, 4.5 K) | $1.6\times10^5$ A/cm$^2$ | $3.2\times10^6$ A/cm$^2$ |
| Derived parameters | | |
| $H_{c2}(0)$ | 140 T | 29 T |
| $\xi(0)$ | $\xi_z=7$ Å | $\xi_0=34$ Å |
| $H_c(0)$ | 2.7 T | |
| $\lambda_{GL}(0)$ | $\lambda_z=1250$ Å | $\lambda_0=260$ Å |
| $\kappa$ | $\kappa_z=37$ | $\kappa_0=7.6$ |

We next consider to what extent the anisotropic properties of $Y_1Ba_2Cu_3O_{7-x}$ can be understood in terms of existing theories. Above 78 K, the linearity of our data indicates agreement with anisotropic Ginzburg-Landau theory. Our experimental results indicate $H_{c2}$ anisotropies of 5:1 near $T_c$ and $H_{c1}$ anisotropies of at least 1:10 at low temperatures. Existing theories cannot account for the greater anisotropy measured in $H_{c1}$. Lawrence and Doniach's results[10] for $H_{c1}$ anisotropy according to both an anisotropic Ginzburg-Landau formulation and a Josephson-coupled layered superconductor model indicate that the anisotropy of $H_{c1}$ should be reciprocal to that of $H_{c2}$ and smaller in magnitude. Kogan and Clem[14] and Kogan[15] and, recently, Balatskii, Burlachkov, and Gor'kov[16] calculate that the anisotropy in $H_{c1}$ should be exactly reciprocal to the $H_{c2}$ anisotropy. If these relationships remain valid down to low temperatures, the larger anisotropy we observe for $H_{c1}$ at low temperatures indicates that the $H_{c2}$ anisotropy at low temperature must be larger than it is near $T_c$. The Klemm-Luther-Beasley[11] extensions of the Josephson-coupled layer model indicate that pronounced upward curvature in $H_{c2}^{\parallel}$ (and therefore an increase in anisotropy) can occur in layered systems when the coherence length, $\xi_z$, becomes small enough that vortex cores fit between the layers. This behavior has been seen in a number of intercalated layered superconductors. While this theory does provide a mechanisms to explain an increase of anisotropy with lowering temperature, our estimates indicate that the vortex core size does not shrink small enough with decreasing temperature to force a crossover to two-dimensional behavior and thus an increase in anisotropy. Some other mechanism is necessary to cause an increase of $H_{c2}^{\parallel}$ and anisotropy. A break in the temperature dependence of $H_{c2}^{\perp}$, like that at 78 K, is not predicted by either the anisotropic Ginzburg-Landau theory or the Josephson-coupled layered theory. One possible cause for this behavior could be a complicated Fermi surface as described by Dalrymple and Prober[7] for $Nb_{1-x}Ta_xSe_2$. Another possible explanation could be a transition from $d$-wave coupling to a combination of $s$- and $d$-wave coupling as has been suggested by Kotliar.[17]

In conclusion, we have established from upper-critical-field data that the high-temperature superconductivity in $Y_1Ba_2Cu_3O_{7-x}$ is three dimensional in nature and in accord with the expectations of anisotropic Ginzburg-Landau theory. The strong anisotropy (5:1) is associated with the Cu-O planes. The larger anisotropy measured for the lower critical field and the temperature dependence of the upper critical field at lower temperatures indicate that, at least, elaborations of the simplest theoretical models for anisotropic superconductors are needed to describe $Y_1Ba_2Cu_3O_{7-x}$ at lower temperatures.

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