RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 38, NUMBER 7 1 SEPTEMBER 1988

Angular dependence of the upper critical field of $Bi_{2.2}Sr_2Ca_{0.8}Cu_2O_{8+\delta}$

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(Received 9 May 1988)

We have measured the electrical resistivity of $Bi_{2.2}Sr_2Ca_{0.8}Cu_2O_{8+\delta}$ in the vicinity of T_c for various angles between the $[CuO_2]_\infty$ double layers of the crystal and the magnetic field. Defining T_c at the transition midpoints, we have measured values for $-dH_{c2}^{\parallel}/dT = 45$ T/K and $-dH_{c2}^{\perp}/dT = 0.75$ T/K for the magnetic field parallel and perpendicular to the $[CuO_2]_\infty$ planes, respectively. This results in an anisotropy of a factor 60. The numerical results are sensitive to the definition of T_c and larger values for the anisotropy cannot be excluded. The results are compared with the anisotropic three-dimensional Ginzburg-Landau theory.

The common presence of the $[CuO_2]_\infty$ planes not only in $(La,Sr)_2CuO_4$ (Ref. 1) and $Ba_2YCu_3O_7$,[2] but also in the recently discovered bismuth[3] and thallium[4] compounds, strongly suggests that the superconductivity originates from these $[CuO_2]_\infty$ planes. Therefore, one might expect a dimensional crossover to two-dimensional superconductivity if the coherence length along the c axis, ξ_c, becomes smaller than the $[CuO_2]_\infty$ layer separation.[5] In fact, the separation between the $[CuO_2]_\infty$ planes in $(La,Sr)_2CuO_4$ and $Ba_2YCu_3O_7$ seems insufficiently large to give rise to two-dimensional superconductivity, in spite of an extremely short coherence length. Experiments probing the anisotropy of $Ba_2YCu_3O_7$ (Ref. 6) are well described by the three-dimensional anisotropic Ginzburg-Landau (GL) theory, which incorporates the anisotropy into the quasiparticle effective mass. Two-dimensional superconductivity might be more likely in the recently discovered[3] compound $Bi_{2.2}Sr_2Ca_{0.8}Cu_2O_{8+\delta}$, because of a much larger $[CuO_2]_\infty$ double-layer separation of about 12 Å,[7] and, as we will show below, an even smaller value of the coherence length ξ_c than that found for $Ba_2YCu_3O_7$.

In search of two-dimensional superconducting behavior or a dimensional crossover, we have investigated the angular dependence of the electrical resistivity of $Bi_{2.2}Sr_2Ca_{0.8}Cu_2O_{8+\delta}$ in a magnetic field. Near T_c we expect the sample in the three-dimensional Ginzburg-Landau regime because of the divergence of the coherence length, ξ, at T_c. We anticipate a dimensional crossover in the temperature regime where the coherence length, $\xi_c(T) = \xi_c(0)(1-T/T_c)^{-1/2}$, is comparable to the $[CuO_2]_\infty$ double-layer separation. Such a dimensional crossover, previously observed in intercalated compounds[8] and metallic multilayers,[9] has as its signature a strong temperature dependence of the ratio $H_{c2}^{\parallel}/H_{c2}^{\perp}$, or deviations in the angular dependence of $-dH_{c2}/dT$ from the predictions of the anisotropic Ginzburg-Landau theory.

Defining T_c at the conventional 50% value of the extrapolated normal-state resistivity, yields a good correspondence for the angular dependence of H_{c2}' with the predictions of GL theory. This definition results in a large value of 60 for the anisotropy of H_{c2}' for the magnetic field parallel and perpendicular to the $[CuO_2]_\infty$ double layers. Still, larger values of the anisotropy cannot be excluded, as a result of limited angular resolution of 0.5°. If we define T_c as the temperature at which the linear regime of $\rho(T)$ extrapolates to $\rho = 0$, we obtain an anisotropy in $H_{c2}' = -dH_{c2}/dT$ of 25. But if we choose this definition, the angular dependence of H_{c2}' is sharper than expected from GL theory.

The high-quality single crystal used in this investigation was grown from an alkali chloride flux, as described by Schneemeyer *et al.*[10] Extensive characterization of these crystals has been described elsewhere.[10] An optically flat piece of a crystal was obtained by cleaving the crystal along the a and b axis to a rectangle of 2.1×0.64 mm^2. We estimate the thickness of the crystal to be between 1 and 2 μm.

Four Ag contacts were sputtered on the crystal in ~0.2-mm strips spanning the entire width of the sample resulting in a conventional bar-shaped geometry. The distance between the voltage contacts was 0.72 mm. The contact resistance was about 2 Ω at room temperature. The sample was glued with a minute amount of GE varnish onto a sapphire substrate and four 25-μm-diam. Ag wires were attached with Ag epoxy.

Measurements were performed in a quick-insert cryostat, with the insert warmed to ambient temperature while changing the angle between the crystal and the magnetic field. The angles have a relative accuracy of 0.5° and we defined the angle $\phi = 0$ for the run with the highest value of H_{c2}'. During the experiments, the room-temperature resistance increased less than 3%.

The resistance was measured using a dc current of 0.5 mA perpendicular to the magnetic field for all angles. The temperature was measured with a calibrated Pt thermometer, and accurate corrections for its magnetoresistance were performed. A magnetic field up to 10 T was applied using a superconducting solenoid.

In Fig. 1, we show the temperature dependence of the electrical resistivity up to room temperature in zero magnetic field. Above 150 K, the normal-state resistivity is linear and fits $R_n = 0.385 + (5.307 \times 10^{-3})T$. Assuming a sample thickness of 1 μm, we obtain a room-temperature resistivity $\rho_n(300\text{ K}) = 220\ \mu\Omega$ cm. Below 150 K, there is pronounced rounding of the resistance curve, continuously progressing into the superconducting transition, although the transition of $\rho(T)$ near $\rho = 0$ is very sharp.

Figure 2 shows the resistive transitions when applying a magnetic field $H = 0$, 2, 5, and 10 T for three different an-

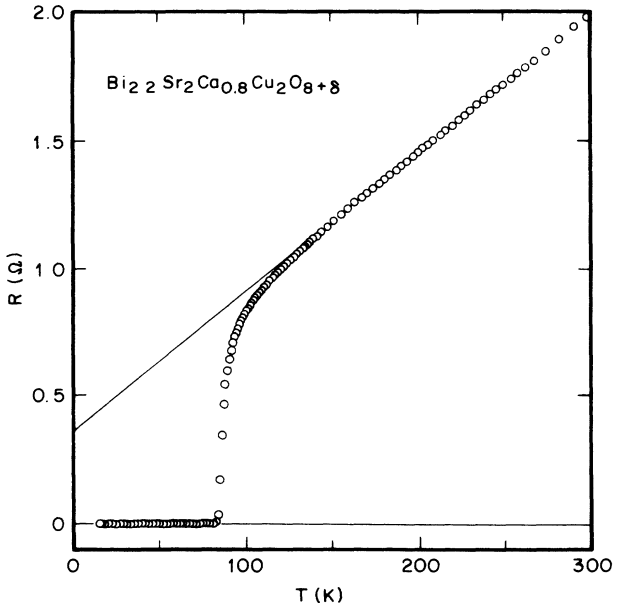

FIG. 1. Temperature dependence of the electrical resistivity of $Bi_{2.2}Sr_2Ca_{0.8}Cu_2O_{8+\delta}$.

gles $\phi=0°$, 4.5°, and 35°. Unfortunately, it is impossible to define an onset temperature of the superconducting transition, even in zero magnetic field. Still, this figure clearly shows that the sample becomes more resistive with increasing magnetic field, and with increasing angle ϕ from $\phi=0$, where the $[CuO_2]_\infty$ double layers are parallel to H, to $\phi=90°$.

To determine the angular dependence of $H'_{c2}=-dH_{c2}/dT$, we *defined* T_c in two ways. First, T_c was defined as the temperature at which $\rho=0$, as extrapolated from the linear regime of $\rho(T)$ in the transition. An example is shown in Fig. 2 for $\rho(H=2\text{ T}, \phi=35°)$. The resulting $H_{c2}(T)$ phase diagram is shown in the inset of the upper part of Fig. 3 for various angles ϕ, exhibiting a concave upturn of $H_{c2}(T)$. This upturn is probably not caused by dimensional crossover, but is likely due to the onset of flux-flow resistivity. The main upper part of Fig. 3 shows the angular dependence of H'_{c2} defined as $5/[T_c(5\text{ T})-T_c(10\text{ T})]$. The resulting anisotropy of H'_{c2} is $H'_{c2}(\phi=0)/H'_{c2}(\phi=90°)=9.1/0.37=25$. For comparison we show the curves as calculated from the anisotropic GL theory

$$H'_{c2}(\phi)=H'_{c2}(90)(\cos^2\phi+\varepsilon^2\sin^2\phi)^{-1/2}$$

for $\varepsilon[=\xi_c/\xi_a=(m_a/m_c)^{1/2}]=0.001$, 0.040, and 0.100, fixing $H'_{c2}(\phi=90°)=0.37$ T/K, as measured. Clearly, the measured points do not fit the GL expression but have a more peaked angular dependence. Also the slopes H'_{c2} between 2 and 5 T or 0 and 2 T yield a more peaked angular dependence than expected from the GL expression.

The temperature at which the resistance has half the value of the extrapolated normal-state resistivity was used for the second definition of T_c, as shown in Figs. 1 and 2. The resulting $H_{c2}(T)$ phase diagram is shown in the inset of the lower part of Fig. 3 for various angles ϕ, resulting in convex curves for $H_{c2}(T)$. Using the same definition for H'_{c2} as above, we obtain the angular dependence of $H'_{c2}(\phi)$

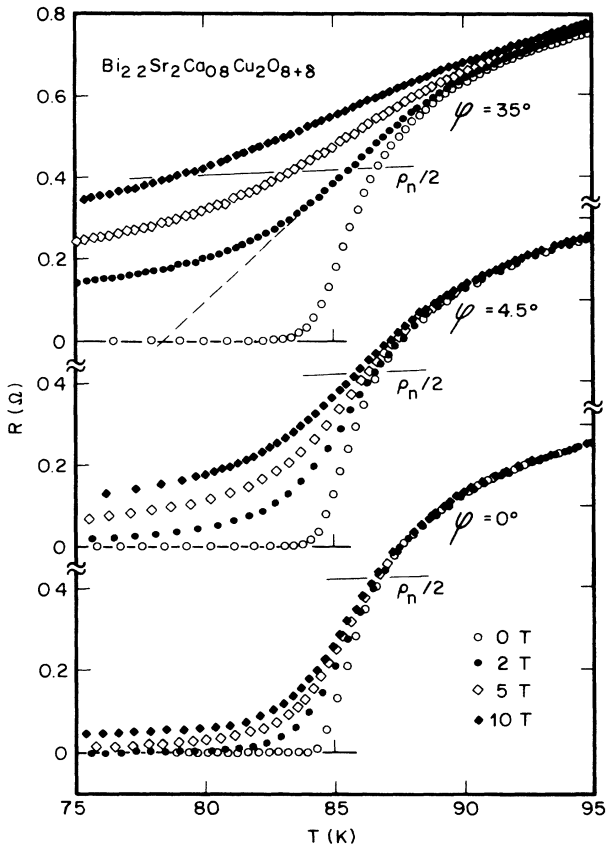

FIG. 2. Temperature dependence of the electrical resistivity of $Bi_{2.2}Sr_2Ca_{0.8}Cu_2O_{8+\delta}$ in magnetics fields of 0, 2, 5, and 10 T for three angles ϕ between the magnetic field and the $[CuO_2]_\infty$ planes. Also indicated are the half values of the extrapolated normal-state resistivity, and one example of the extrapolation to $\rho=0$ for $H=2$ T at $\phi=35°$.

shown in the lower part of Fig. 3. Again for comparison three anisotropic GL curves are shown, now for $\varepsilon=0.001$, 0.014, and 0.020 fixing $H'_{c2}(90°)=0.75$ T/K. The anisotropy resulting from this definition of T_c is $H'_{c2}(\phi=0°)/H'_{c2}(\phi=90°)=45/0.75=60$, but the GL curves clearly show that an even larger anisotropy may be possible.

Before addressing the anisotropy of the $H_{c2}(\phi)$ data, it is appropriate to discuss the resistive behavior in the vicinity of T_c. Figure 2 shows that there is considerable magnetoresistance in the onset of the superconducting transition. The data extend to higher temperatures and show that the ρ-T curves for different values of the magnetic field do not give a distinct resistive anomaly at the temperature where the magnetoresistance vanishes, but rather the $\rho(T)$ curves smoothly fan out at $\sim$105 K. The rounding of $\rho(T)$ starts at even higher temperatures ($\sim$140 K).

We suggest two sources for the magnetoresistance and the rounding of $\rho(T)$. First, the crystal may contain intergrowths, resulting in local inhomogeneities of higher T_c phases of the Bi-Sr-Ca-Cu-O system.[11] Second, the deviations from linearity of $\rho(T)$ can be the result of fluctuations,[12] which can extend to temperatures far above T_c in the high-T_c superconductors. A combination of both effects is also possible. We addressed the possibility of

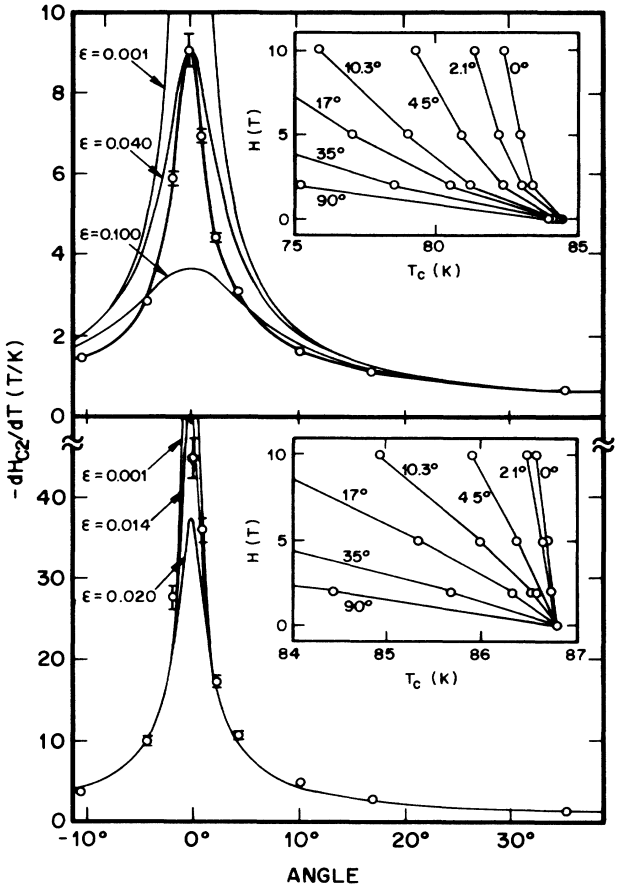

FIG. 3. Angular dependence of the upper critical-field slope of $Bi_{2.2}Sr_2Ca_{0.8}Cu_2O_{8+\delta}$. The upper part of the figure uses the extrapolated zero-resistivity definition of T_c, and the lower part uses the transition midpoints. The thick line in the upper part is a guide to the eye through the data points. The thin lines are GL curves for three values of ε. The insets show the corresponding $H_{c2}(T)$ phase diagram.

fluctuations by fitting the excess conductivity $\sigma'(T)$, defined as the difference between the extrapolated normal-state values and the measured conductivity, to a power law: $\sigma' \propto (T/T_c - 1)^{\alpha}$. Although this power law fits the data very well, the accuracy in determining T_c is insufficient for the determination of $H'_{c2}(T)$. Furthermore, we obtained large negative values for α, $(-3 < \alpha < -1)$, which is inconsistent with the appropriate theories of direct or indirect contributions to the conductivity due to fluctuations.[12] To determine a possible contribution of inhomogeneities to $\rho(T)$ above 86 K, we tried to determine the normal-state resistivity from the functional dependence of $\rho = \rho(H)$ and then taking the limit for $H \rightarrow \infty$ or $H \rightarrow H_{c2}$. This attempt was unsuccessful and thus we have not been able to separate both contributions. We expect however, that inclusions of a 105-K phase should have extremely high critical-field values in the temperature regime where we extract our values of T_c and therefore would not appreciably affect the resistance drop at 85 K.

Below T_c the $\rho(T)$ behavior is also complicated. First, we found that the pinning force must be very low as no change in the resistivity was found upon varying the current by four orders of magnitude between 0.5 μA and 5 mA, between 60 and 80 K for $\phi = 90°$ and $H = 2$ T. Also, the flux-flow resistance ρ_f does not follow the rule[13] $\rho_f/\rho_n = H/H_{c2}(0)$. Instead, $d\rho_f/dH$ decreases significantly for relatively low fields (compared to an estimate of H_{c2}), but does not saturate below 10 T.

These results led us to choose two definitions of T_c. First, we used an extrapolation scheme of $\rho(T)$, as shown in Fig. 2 for $\rho(H = 2\text{ T}, \phi = 35°)$. This scheme was successfully used for $Ba_2YCu_3O_7$, where inhomogeneities can give rise to a "resistance foot," preventing the definition of T_c at the value where $\rho(T) = 0$. Better data for single-crystal data justified the use of this procedure,[14] although this definition for T_c results in a low value of T_c and a lower limit to the anisotropy. Second, we used the more conventional definition of T_c at the arbitrary value $\rho(T) = 0.5\rho_n(T)$, extrapolating ρ_n from the high-temperature behavior. We note that flux flow sets in below $0.5\rho_n(T)$ and thus criteria using smaller values of $\rho(T)/\rho_n(T) = 0.15$ and 0.30 as reported by Juang *et al.*[15] give results that are influenced by this flux-flow resistance.

Using these definitions for T_c, we see that the first choice suggests an anisotropy of H'_{c2} of 25, but fails to give agreement with the angular dependence predicted by the anisotropic GL theory. The second choice yields an anisotropy of H'_{c2} of 60 or even larger, and the data points are in agreement with GL theory, as expected (see below). We emphasize that anisotropy values larger than 60 fit the data equally well as clearly shown in Fig. 3. From this plot it is obvious that the anisotropy we measured could be limited by the experimental angular resolution. In contrast to these results on $Bi_{2.2}Sr_2Ca_{0.8}Cu_2O_{8+\delta}$, results for $Ba_2YCu_2O_7$ show that both T_c definitions gave results consistent with the GL angular dependence.[6] For $Ba_2YCu_3O_7$ the zero-resistance criterion yielded an anisotropy in H'_{c2} of 3.0 and the midpoints yielded a value of 4.7. Apparently the anisotropy of $Bi_{2.2}Sr_2Ca_{0.8}Cu_2O_{8+\delta}$ is much larger than that observed for $Ba_2YCu_3O_7$. The deviations from GL theory, obtained using the zero-resistance definition, are most likely due to the large flux-flow resistance for large values of the angle ϕ.

The angular dependence of H'_{c2} implies that extremely good alignment of the crystal is required to obtain reliable data. An error of 2° reduces the value of $H'^{\parallel}_{c2}$ by about a factor 2. Using our values of $H'^{\parallel}_{c2}$ as conservative estimates, we obtain, using the Werthamer-Helfand-Hohenberg theory,[16] values for $H^{\perp}_{c2}(0) = 22(44)$ T and $H^{\parallel}_{c2}(0) = 533(2640)$ T for the zero-resistance and midpoint definition, respectively. From these values we calculate zero-temperature coherence lengths $\xi_a(0) = 38(27)$ Å and $\xi_c(0) = 1.6(0.45)$ Å. This is in agreement with our earlier conclusion that $\xi_c(0)$ becomes smaller than the $[CuO_2]_\infty$ double-layer separation[17] leading us to expect the dimensional crossover at that temperature at which $\xi_c(T)$ equals the double-layer separation.

We can estimate the temperature of the dimensional crossover from our measurements, where $\xi_c(T) = s/\sqrt{2}$ with s the interlayer spacing: $s \approx 12$ Å. By calculating the coherence length ξ_a from $\xi_a^2(T) = \phi_0/2\pi H^{\perp}_{c2}(T)$ and ξ_c from $\xi_a(T)\xi_c(T) = \phi_0/2\pi H^{\parallel}_{c2}(T)$, we estimate the di-

mensional crossover for $\xi_c^2(T)=(\phi_0/2\pi)\ (H_{c2}^{\perp}/H_{c2}^{\parallel})\times(1/H_{c2}^{\parallel})=s^2/2$. Using our value of the anisotropy of $H_{c2}^{\parallel}/H_{c2}^{\perp}=25(60)$, we expect the dimensional crossover for $H_{c2}^{\parallel}\approx 17(7)$ T. This means that the dimensional crossover should be on the borderline of our experimental regime, and accessible experimentally. If the anisotropy we have measured is indeed not intrinsically limited, but limited by the angular alignment of the crystal, the required magnetic fields would be even smaller. In this case it is possible that we have already entered the two-dimensional regime accounting for the high value of $H_{c2}^{\parallel\prime}$. However, to observe the dimensional crossover as an upward curvature in the 45 T/K slope $H_{c2}^{\parallel\prime}(T)$ remains an experimental challenge.

Finally, we want to compare the value of the anisotropy of the upper-critical fields of 25(60) with other anisotropies. First, the Fermi velocity anisotropy, derived from calculated band structures,[18] is approximately 13. In the clean limit this would result in an H_{c2} anisotropy of ~ 3.6. The present experimental values are an order of magnitude larger and thus point to additional contributions, such as mean-free-path anisotropies. Secondly, resistivity measurements by Martin *et al.*[19] yielded resistivity anisotropy values of the order of 10^5, which is at least two orders of magnitude larger than expected from our value of the H_{c2} anisotropy. We note, however, that these large values of $\rho_{\perp}/\rho_{\parallel}$ might be, at least partly, due to the nature of the samples: they contain various intergrowth layers some of which might be insulating. These sample imperfections affect the macroscopic current flow along the c axis. In contrast, the upper-critical field is associated with microscopic currents on a length scale given by the vortex size. Therefore the present H_{c2} anisotropy is intrinsic to the superconductor.

In conclusion, our measurements indicate a large anisotropy of the superconducting properties of $Bi_{2.2}Sr_2Ca_{0.8}Cu_2O_{8+\delta}$. Depending on the definition of T_c we find an anisotropy of H_{c2}' of 25 or 60 and even larger values for the anisotropy cannot be excluded. In spite of the deviations from anisotropic Ginzburg-Landau theory, a clear dimensional crossover has not been observed. Our results indicate that the magnetic fields required to observe a dimensional crossover should be experimentally accessible. It is clear that attempts to measure the anisotropy of $Bi_{2.2}Sr_2Ca_{0.8}Cu_2O_{8+\delta}$ will be seriously complicated by the large flux-flow resistance which makes the determination of $T_c(H)$ very difficult.

We wish to acknowledge stimulating discussions with S. Martin, A. T. Fiory, A. F. Hebard, and A. P. Ramirez. We also thank L. F. Mattheiss for providing us with the calculations of the Fermi velocity anisotropy. P. Marsh and R. M. Fleming performed the x-ray analysis. The computer fitting program was written by G. J. Nieuwenhuys.

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