Upper critical field, fluctuation conductivity, and dimensionality of $YBa_2Cu_3O_{7-x}$

B. Oh, K. Char, A. D. Kent, M. Naito, M. R. Beasley, T. H. Geballe, R. H. Hammond, and A. Kapitulnik
Department of Applied Physics, Stanford University, Stanford, California 94305

J. M. Graybeal
Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139
(Received 4 December 1987)

The upper critical field H_{c2} and fluctuation conductivity were measured for highly oriented thin films of $YBa_2Cu_3O_{7-x}$. The H_{c2} results demonstrate the intrinsic anisotropy in this layered superconductor. The broadening of the resistive transition under fields is interpreted in terms of critical fluctuations. The fluctuation conductivity shows dimensional crossover as expected for quasi-two-dimensional material. Using these data we determine the intralayer and interlayer coherence lengths $\xi_c(0)$ and $\xi_{ab}(0)$, and discuss the dimensionality of the superconductivity in this material.

The recent discovery of high-T_c superconductivity in metallic oxides has generated great interest in the nature of the superconductivity in these materials.[1,2] Since the crystal structure is strongly anisotropic, the transport properties are expected to depend on the crystal orientation. Indeed, their anisotropic nature has been observed in resistivity,[3] critical-current-density,[4] and upper-critical-field measurements.[5-8] Similarly, the fluctuations above the superconducting transition are expected to reflect the degree of anisotropy.

In this paper, we analyze the upper critical field, the marked broadening of the resistive transition in a field and the fluctuation conductivity above T_c for highly oriented thin films of $YBa_2Cu_3O_{7-x}$. With the hypothesis of strong thermodynamic fluctuations, we can self-consistently account for all these properties and demonstrate that this material is distinctly quasi-two-dimensional with the superconductivity existing in the Cu-O (a-b) planes (possible also in the chains) and with Josephson coupling between planes.

In these experiments, we used polycrystalline epitaxial thin films of $YBa_2Cu_3O_{7-x}$ with thicknesses $\sim 1\ \mu m$ grown on $SrTiO_3(100)$ substrates by electron-beam evaporation and magnetron sputtering. Preparation and characterization of the samples were reported elsewhere.[9-11] The resistive transitions were measured using a four-point probe dc method and the data used to determine both the upper critical field $H_{c2}(T)$ and the fluctuation conductivity as described in detail below. For these resistance measurements, the samples were patterned and chemically etched into 400-μm-wide strips by standard photolithographic methods. To get a good contact, the surface is ion-milled, and then 100 Å of titanium and 1000 Å of silver were evaporated sequentially on the films to form contact pads; indium was then pressed onto these pads to form the actual contact.

Figures 1(a) and 1(b) depict the zero-field and high-field resistive transitions of a typical c-axis oriented (the c axis is perpendicular to the film plane) sample 1. The line indicates the resistive transition measured by a temperature sweep at a fixed field and the solid points by a field sweep at a fixed temperature. The resistive transition region clearly broadens as the applied field increases. One striking effect evident in these data and evident in the data of many groups[5,8] is that as a magnetic field is applied, the transition broadens dramatically but with only a small effect in the region near the onset of the transition.

For homogeneous conventional type-II superconductors, the determination of H_{c2} as a function of temperature is usually not sensitive to the criterion used to define T_c. This is because the resistive transitions do not dramatically change their shape as the field increases. The customary choice of convenience for T_c has been the point where the resistance drops to half of its normal-state value. An important starting point for understanding the superconducting transitions of this oxide superconductor $YBa_2Cu_3O_{7-x}$ is to understand the difference between its transitions and those of conventional superconductors. We propose below two alternative explanations, of which the second is more attractive in our judgement.

One possible explanation for the observed broadening invokes inhomogeneities and strong disorder. In this interpretation, as a magnetic field is applied, vortex pinning

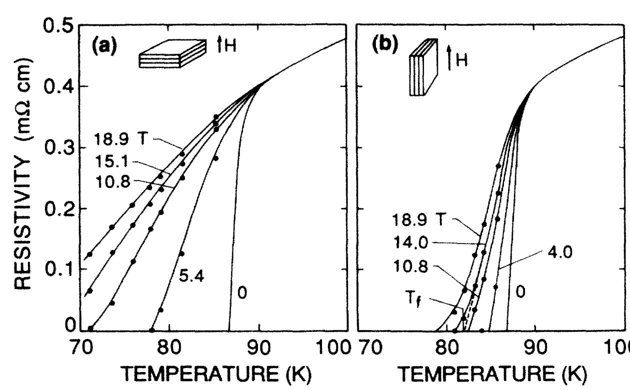

FIG. 1. Temperature dependence of resistivity of sample 1 in fields (a) perpendicular, and (b) parallel to the Cu-O planes. T_c defined by linear extrapolation of the resistivity to $\rho = 0$ is shown in (b).

at the weak parts of the sample degrades rapidly, reducing the connectivity of the material and resulting in a strong broadening of the transition. If this were the case, then the upper (i.e., higher temperature) part of the transition is more representative of the pure material's upper critical field. Taking this point of view, we can estimate $H_{c2}(T)$ by defining T_c as the temperature at which ρ is 90% of its extrapolated normal value ($0.9\rho_N$ criterion). The shape of the $H_{c2}(T)$ phase boundary determined this way is a straight line. The Cu-O planes in c-axis oriented films are parallel to the film plane so that we could readily estimate both H_{c2}^{c} and H_{c2}^{ab}. The coherence lengths along the c axis $\xi_c(0)$ and in the ab plane $\xi_{ab}(0)$ can be estimated from the anisotropic Ginzburg-Landau relations.[12]

For the above choice of T_c ($0.9\rho_N$ criterion), we find $\xi_c(0)\sim 2$ Å and $\xi_{ab}(0)\sim 13$ Å. Since the H_{c2}^{ab} for c-axis oriented samples has been measured with the field parallel to the thin-film plane, there was a possibility that the results might be affected by surface superconductivity. To check this, we also measured H_{c2}^{ab} for an a-axis oriented sample using the $0.9\rho_N$ criterion. The field is perpendicular to the film plane in this case, and there is no surface superconductivity. The result is very similar to the H_{c2}^{ab} of the c-axis oriented films. Thus, our results appear free of the effects of surface superconductivity.

The short coherence lengths obtained by this analysis imply that critical fluctuations may be important and suggest a second interpretation of the observed broadening in H_{c2}. If critical fluctuations occur, it is known that

$$H_{c2}(T)=\Phi_0/2\pi\xi(T)^2 = [\Phi_0/2\pi\xi(0)^2]\{[T_c(0)-T_c(H)]/T_c(0)\}^{2\nu} ,$$

where ν is the coherence length exponent that is rigorously greater than the mean-field value $\nu=\frac{1}{2}$.[13,14] Assuming that the broadening in the field is only due to critical fluctuations and that T_c is given by a linear extrapolation of the resistivity to $\rho=0$ as shown in Fig. 1(b), we test this hypothesis as follows. Fitting H_{c2}^{c} to the above formula as in Fig. 2, we find $\nu=0.65\pm0.02$ and $\xi_{ab}(0)=16\pm2$ Å for three different samples: sample 2 made by electron-beam evaporation, sample 3 made by magnetron sputtering, and the single-crystal data of Iye *et al.*[8] Hence, the exponent ν appears to be universal. The same plot for

$$H_{c2}^{ab}(T)=\Phi_0/2\pi\xi_c(T)\xi_{ab}(T) = [\Phi_0/2\pi\xi_c(0)\xi_{ab}(0)]\{[T_c(0)-T_c(H)]/T_c(0)\}^{2\nu}$$

shows similar behavior except that the exponent ν is found to be 0.80 ± 0.07. Also, taking $\xi_{ab}(0)=16$ Å from the H_{c2}^{c} data, we find $\xi_c(0)=2.2\pm1.0$ Å. The fact that the coherence lengths determined using the $0.9\rho_N$ criterion agree with the values determined assuming critical fluctuations likely reflects that the upper part of the transition is further from the critical region and thus approaches the mean-field value for ν.

Let us now turn to the question of the dimensionality of these superconductors and then to their fluctuation conductivity. Since $\xi_c(0)$ is smaller than any separation between Cu-O planes, $YBa_2Cu_3O_{7-x}$ is a quasi-two-dimensional system, and $\xi_c(0)$ is related to the interlayer cou-

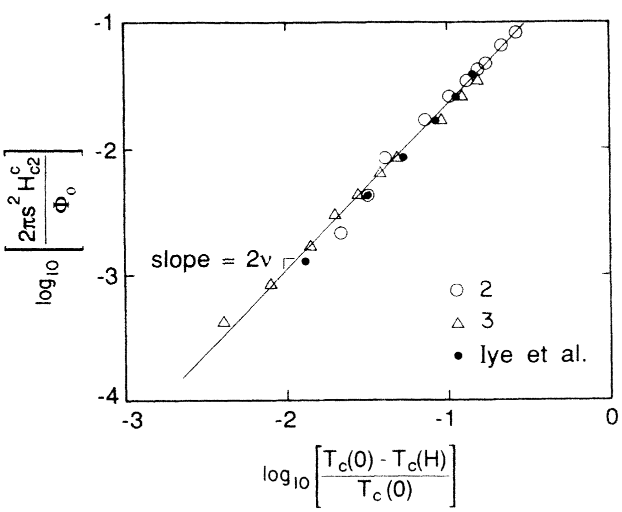

FIG. 2. Test for critical fluctuations from H_{c2}^{c} data of three samples: two c-axis-oriented thin films (samples 2 and 3) measured by us and the single-crystal data of Iye *et al.* (Ref. 8). The periodicity $s=11.7$ Å was used. The data yield a critical exponent $\nu=0.65\pm0.02$ and an in-plane coherence length $\xi_{ab}(0)=16\pm2$ Å as described in the text.

pling. If $YBa_2Cu_3O_{7-x}$ is quasi two dimensional, dimensional crossover effects should be present. Unfortunately, only a small temperature range below T_c can be covered by measuring the upper critical field of $YBa_2Cu_3O_{7-x}$ because of the rapid increase in H_{c2} as T is reduced. As a result, no crossover in H_{c2} data was observed. Moreover, it is not clear what would be the influence of critical fluctuations on this crossover. On the other hand, measurement of the fluctuation-enhanced conductivity σ' can cover a large temperature range above T_c. In addition, if we are far enough above T_c, mean-field theory should apply, and we can use standard formulas to fit the fluctuation conductivity. The fit will of course have to be restricted to outside the critical region. In addition, fluctuation conductivity measurements provide a complementary way to determine $\xi_c(0)$ and to establish the dimensionality of $YBa_2Cu_3O_{7-x}$. Only c-axis oriented samples were measured to make sure that the current flows mainly along Cu-O planes. The deviation of the measured resistivity from $\rho_N(T)$, defined as $\Delta\rho(T)$, measures the excess conductivity $\sigma'(T)$. Simple algebra yields $\sigma'(T)=\Delta\rho(T)/\rho(T)\rho_N(T)$. Outside the critical region, $\sigma'(T)$ is a function of $\varepsilon=(T-T_c^{\mathrm{mf}})/T_c^{\mathrm{mf}}$ only, where T_c^{mf} is the mean-field transition temperature, hence the determination of T_c^{mf} is of primary importance to the determination of $\sigma'(T)$. The T_c^{mf} was determined by extrapolating the linear three-dimensional (3D) region of a σ'^{-2} vs T plot as in the inset of Fig. 3, since as $T\rightarrow T_c^{\mathrm{mf}}$ σ' should diverge as $(T-T_c^{\mathrm{mf}})^{-1/2}$. Note that $T_c^{\mathrm{mf}}>T_c$ and typically the shift is of $1\sim2$ K, which is a rough measure of the size of the critical region above T_c. This is self-consistent with our assumption of critical fluctuations close to T_c and mean-field behavior above $T_c^{\mathrm{mf}}>T_c$. The data in Fig. 2 imply that the critical region below T_c is much larger than the one above T_c. This result is contrast

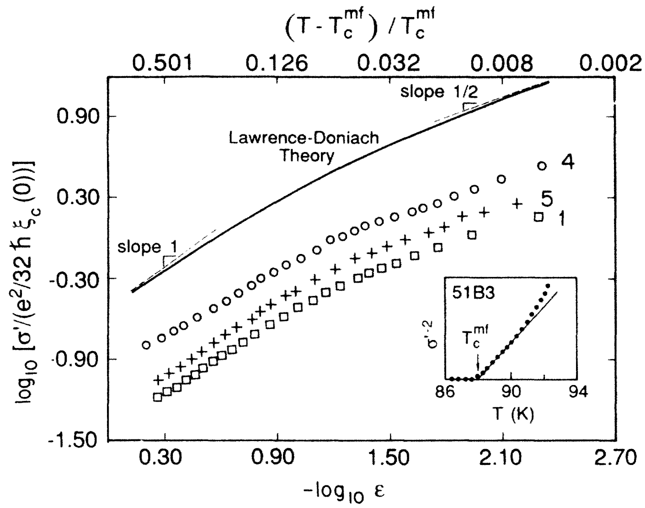

FIG. 3. Fluctuation conductivity above T_c^{mf} for samples 1, 4, and 5 compared with the Lawrence-Doniach (LD) theory. The results for $YBa_2Cu_3O_{7-x}$ thin films show 2D to 3D crossover in qualitative accord with theory. In plotting this figure, $\xi_c(0)$ was taken as 1.85 Å. The inset shows σ'^{-2} vs T to determine T_c^{mf}.

to the prediction of the *inverted XY* model by Dasgupta and Halperin.[15]

Figure 3 shows values of σ' for several of our samples compared with the prediction of Lawrence-Doniach (LD) theory. Within the LD theory the fluctuation-enhanced conductivity in the *a-b* plane is [12]

$$\sigma'(\varepsilon)=e^2\{1+[2\xi_c(0)/s]^2\varepsilon^{-1}\}^{-1/2}/16\varepsilon s\hbar \ , \tag{1}$$

where $\varepsilon=T/T_c^{mf}-1$ and s is the layer periodicity. In this equation we note that σ' will diverge as $\varepsilon^{-1/2}$ (3D behavior) when the temperature is close to T_c^{mf}, and that σ' will go as ε^{-1} dependence (2D behavior) at sufficiently high temperature such that $2\xi_c(T)/s<1$. The dimensional crossover temperature is

$$T_0=T_c^{mf}\{1+[2\xi_c(0)/s]^2\} \ . \tag{2}$$

Since the relevant temperature range is far from T_c^{mf} and inelastic scattering seems to be important in this material,[16] the Maki-Thompson contribution[17,18] was not considered. Moreover, since the coherence lengths are very short [$\xi_c(0)$ is comparable to atomic distances], there is no theoretical need to consider a high-energy cutoff.

A 2D-to-3D crossover behavior is evident in accord with the LD theory in Fig. 3. The dimensional crossover temperature T_0 for every sample determined from this plot is listed in Table I. The LD theory curve was calculated assuming $T_c=1.1T_c^{mf}$, which is the average value observed for the three samples. In Fig. 3, we note that the experimental data lie below the theory by a factor $1/C$, which is different for each sample. This C factor for each sample is also listed in Table I. One possible origin for this scaling factor is that the current flow in the film is still not uniform on a submacroscopic scale, due perhaps to poor grain boundaries, microcracks, or uneven oxidation, in spite of the much improved quality of these epitaxial films.

TABLE I. The results for fluctuation-conductivity measurements and the parameters for LD theory.

Sample	T_c^{mf} (K)	$\frac{T_0}{T_c^{mf}}$	C	$\xi_c(0)$ (Å)	$\frac{d\rho}{dT}$ ($\mu\Omega$ cm/K)	$\frac{1}{C}\frac{d\rho}{dT}$
4	87.2	1.12	3.3	1.9	1.70	0.52
5	84.5	1.05	5.5	1.5	2.93	0.53
1	87.9	1.07	7.4	1.6	4.80	0.64
LD theory		1.1[a]	1.0	1.85		

[a]Average value for the three samples.

In such an event, the resistivity determined by the geometrical dimension of the film would be too large by some factor. It is interesting to note that the derivative of the observed normal-state resistivity $d\rho/dT$ scaled by the observed factor C shows a very close correlation among the samples listed in Table I. This suggests that $d\rho/dT$ scaled by factor C, which we take as the true $d\rho/dT$, is an intrinsic property of this material just as one might expect. In addition, it brings the fluctuation conductivity theory and data into better agreement.

Using the observed T_0, we obtain $\xi_c(0)=1.5\sim 2$ Å from Eq. (2) with $s=11.7$ Å (see Table I). There is some ambiguity about the correct periodicity to determine $\xi_c(0)$. However, since $2\xi_c(0)/s$ depends only on $T_0/T_c^{mf}\sim 1.1$, we obtain directly that $2\xi_c(0)/s\sim 0.3$ for our samples independent of the correct interpretation of s. Thus, in any event, these data show that $YBa_2Cu_3O_{7-x}$ is a quasi-two-dimensional-layered superconductor.

The results of our short coherence lengths and effective dimensionality differ from those of Freitas, Tsuei, and Plasket,[19] Worthington, Gallagher, and Dinger,[6] and Gallaghar *et al.*[7] Freitas *et al.*[19] have examined the fluctuation conductivity for polycrystalline materials with single coherence length. However, since $YBa_2Cu_3O_{7-x}$ has a intrinsic anisotropic nature, single crystals or oriented thin films are preferred to study the effective dimensionality. Worthington *et al.*[6] and Gallagher *et al.*[7] used single crystals and measured $H_{c2}(T)$ inductively. Note that these authors define $\xi_c(0)^2=\Phi_0/[2\pi 0.69T_c(dH_{c2}^c/dT)_{T_c}]$. Using our definition of $\xi_c(0)$, their values become 5.8 and 3.6 Å, respectively. If we arbitrarily take a $\rho/\rho_N=0.5$ criterion as has been commonly done, and assume a linear fit, our data yield $\xi_c(0)\sim 3.8$ Å. However, there is marked curvature at low fields in contrast to the $\rho/\rho_N=0.9$ criterion or as done in Fig. 2.

It is worth examining the implications of the short coherence lengths and the effective two dimensionality of the superconductivity in $YBa_2Cu_3O_{7-x}$. Assuming that the superconducting layers are the Cu-O planes and that there is one carrier per unit cell, the density of the carriers in the unit area $n_{2D}\sim 3.3\times 10^{14}$ cm^{-2}. For an in-plane coherence length of 16 Å, this corresponds to a maximum of four pairs per coherence volume, much less than in conventional superconductors. Such a low number of pairs in a coherence volume is related to the presence of critical fluctuation at the transition. It also implies that within a BCS-like picture most of the Fermi sphere participates in the pairing interaction.

In conclusion, we have argued in this paper that $YBa_2Cu_3O_{7-x}$ exhibits quasi-two-dimensional superconductivity with very short coherence lengths, and that the transition exhibits critical fluctuations with a correlation length exponent of ~ 0.65.

The authors gratefully acknowledge experimental assistance from J. Juang, as well as B. Brandt of the Francis Bitter National Magnet Laboratory (FBNML). A. K. was partially supported by the Alfred P. Sloan Foundation. Part of this work was performed at FBNML, which is supported at MIT by the National Science Foundation (NSF). J. M. G. wishes to thank the AT&T Foundation for financial support. This work was supported by U.S. Air Force Office of Scientific Research, U.S. Office of Naval Research, and NSF through the various participants. Materials were characterized using the facilities of the Center for Materials Research at Stanford University under NSF-Materials Research Laboratory support.

[1] J. G. Bednorz and K. A. Müller, Z. Phys. B **64**, 189 (1986).

[2] M. K. Wu, J. R. Ashburn, C. J. Torng, P. H. Hor, R. L. Meng, L. Gao, Z. J. Huang, Y. Q. Wang, and C. W. Chu, Phys. Rev. Lett. **58**, 908 (1987).

[3] S. W. Tozer, A. W. Kleinsasser, T. Penney, D. Kaiser, and F. Holtzberg, Phys. Rev. Lett. **59**, 1768 (1987).

[4] T. R. Dinger, T. K. Worthington, W. J. Gallagher, and R. L. Sandstrom, Phys. Rev. Lett. **58**, 2687 (1987).

[5] Y. Hidaka, Y. Enomoto, M. Suzuki, M. Oda, A. Katsui, and T. Murakami, Jpn. J. Appl. Phys. **26**, Pt. 2, L726 (1987).

[6] T. K. Worthington, W. J. Gallagher, and T. R. Dinger, Phys. Rev. Lett. **59**, 1160 (1987).

[7] W. J. Gallagher, T. K. Worthington, T. R. Dinger, F. Holtzberg, D. L. Keiser, and R. L. Sandstrom (unpublished).

[8] Y. Iye, T. Tamegai, H. Takeya, and H. Takei, Jpn. J. Appl. Phys. **26**, Pt. 2, L1057 (1987).

[9] B. Oh, M. Naito, S. Arnason, P. Rosental, R. Barton, M. R. Beasley, T. H. Geballe, R. H. Hammond, and A. Kapitulnik, Appl. Phys. Lett. **51**, 852 (1987).

[10] M. Naito, R. H. Hammond, B. Oh, M. R. Hahn, J. W. P. Hsu, P. Rosental, A. F. Marshall, M. R. Beasley, T. H. Geballe, and A. Kapitulnik, J. Mater. Res. **2**, 713 (1987).

[11] K. Char, A. D. Kent, A. Kapitulnik, M. R. Beasley, and T. H. Geballe, Appl. Phys. Lett. **51**, 1370 (1987).

[12] W. E. Lawrence and S. Doniach, in *Proceedings of the Twelfth International Conference on Low Temperature Physics, Kyoto, 1970,* edited by Eizo Kanda (Keigaku, Tokyo, 1971), p. 361.

[13] A. Kapitulnik, M. R. Beasley, C. Castellani, and C. DiCastro, Phys. Rev. B **37**, 537 (1988).

[14] C. J. Lobb, Phys. Rev. B **36**, 3930 (1987).

[15] C. Dasgupta and B. I. Halperin, Phys. Rev. Lett. **47**, 1556 (1981).

[16] P. A. Lee and N. Read, Phys. Rev. Lett. **59**, 2691 (1987).

[17] K. Maki, Prog. Theor. Phys. **39**, 897 (1968); **40**, 193 (1968).

[18] R. S. Thomson, Phys. Rev. B **1**, 327 (1970).

[19] P. P. Freitas, C. C. Tsuei, and T. S. Plaskett, Phys. Rev. B **36**, 833 (1987).