

Upper critical fields of high- T_c superconducting $\text{La}_{2-x}\text{Sr}_x\text{CuO}_{4-y}$: Possibility of 140 tesla

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(Received 2 February 1987)

The upper critical fields $B_{c2}(T)$ of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ ($x=0.15$ and $x=0.20$) were measured near T_c in dc fields to 23 T and at 4.2 K with pulsed fields to 45 T. Resistive midpoint transitions for the $x=0.15$ sample yield $(dB_{c2}/dT)_{T=T_c}=2.2$ T/K, $T_c=38.1$ K, and $B_{c2}(0)=58$ T assuming a dirty type-II superconductor with no Pauli paramagnetic limiting. Data for the onset yield 4.5 ± 0.5 T/K, 40.1 K, and 125 ± 15 T, respectively. Pulsed-field data show that fractions of both materials are superconducting above 45 T at 4.2 K. Calculations of limits of $B_{c2}(T)$ are presented and experimental limitations of measurements in these high-field superconductors are indicated.

Since Bednorz and Müller¹ reported a high T_c in a superconducting oxygen-defect perovskite, many groups have begun investigations in this class of materials (see Refs. 2-6: These are among the papers known to us but there are many other groups actively involved). In this Rapid Communication we report measurements of the upper critical fields, $B_{c2}(T)$, with applied dc fields B_0 up to 23 T (near T_c) and with pulsed fields at 4.2 K up to 45 T in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ for $x=0.15$ and 0.20. We observe that extremely high upper critical fields can be achieved. $B_{c2}(T)$ is a sensitive function of composition and the resistive transition becomes very broad so that very large fields are necessary to define $B_{c2}(T)$. Calculations of $B_{c2}(T)$ are presented to show the wide range of $B_{c2}(0)$ which can be extrapolated based on which assumptions are made. The lowest estimates of $B_{c2}(0)$ are well within our pulsed-field capabilities, whereas the more optimistic projections suggest $B_{c2}(0)=140$ T. The present results are consistent with the assumptions that these new high- T_c materials are conventional superconductors with correspondingly high values of $B_{c2}(T)$.

The materials were prepared at Bell Communications Research as described in Ref. 2 which also discussed structural, magnetic, and electronic properties over the range of $0.05 \leq x \leq 1.1$. The characteristics of the particular samples discussed in this Rapid Communication are given in Table I of Ref. 2 as 2S14 and 4S14 for $x=0.15$ and $x=0.20$, respectively.

The upper critical field $B_{c2}(T)$ was measured resistively with a standard four-probe method in transverse dc magnetic fields up to 23 T furnished by water-cooled magnets at the Francis Bitter National Magnet Laboratory facility. Leads were attached with Ag paint and current densities ≤ 0.5 A/cm² were used. Tests were made to ensure that $B_{c2}(T)$ was not sensitive to the applied dc current. The samples were small rectangular polycrystalline slabs with

good mechanical integrity. The resistive measurements were made in a small cryostat having variable temperature control. The samples were in contact with He gas in the cryostat and the temperature was monitored with capacitance and carbon-glass thermometers. Most of the data were taken at fixed values of T and swept B_0 and selected points were taken at fixed B_0 while varying T . The results of these $B_{c2}(T)$ measurements near T_c are shown in Fig. 1. Detailed data are given for the $x=0.15$ sample because

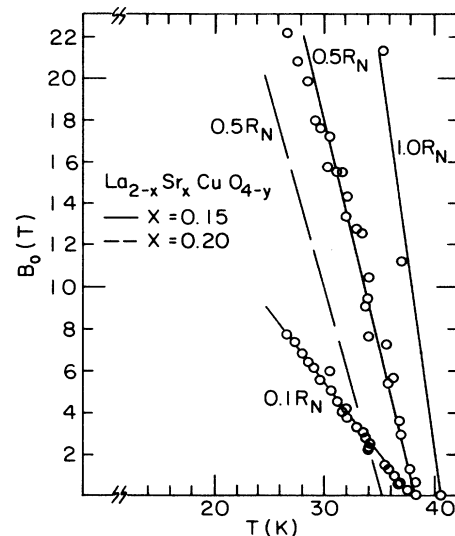


FIG. 1. Upper critical field B_{c2} vs temperature T for $x=0.15$ and 0.20 in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$. The data are corrected for the temperature dependence of the normal resistance. The values of $B_{c2}(T)$ for midpoint resistivity ($0.5R_N$), at 10% ($0.1R_N$), and the onset ($1.0R_N$) are shown for the $x=0.15$, and for midpoint for the $x=0.20$ sample (broken line).

this specimen had the higher T_c and sharper resistive transition at $B_0=0$. The data for the $x=0.20$ sample illustrate the effect of small changes in composition.

There is a rapid increase in the resistive transition width as T is decreased below T_c and this makes it difficult to define the resistive transitions accurately over a large range of temperature unless fields in the range of 20 T or more are available. To illustrate this point we show the data for the resistive midpoint ($0.5R_N$), the bottom of the transition ($0.1R_N$), and the onset which we designate as ($1.0R_N$). In contrast with conventional superconductors, these high- T_c materials show a relatively large linear decrease of R_N near T_c . The data plotted here are corrected for this assumed change in R_N vs T below T_c [which reduces $|(dB_{c2}/dT)_{T=T_c}|$ by about 10% for the $x=0.15$ sample]. A much smaller effect is the magnetoresistance of the material near T_c , which is about 3% at 23 T and nearly T independent in the range we could investigate near T_c . The data taken at fixed B_0 while varying T were consistent with the field-swept data (as expected), and were more convenient for defining the onset of superconductivity.

For the $x=0.15$ sample the midpoint yields $(dB_{c2}/dT)_{T=T_c}=2.2$ T/K and $T_c=38.1$ K; for the onset $(dB_{c2}/dT)_{T=T_c}=4.5 \pm 0.5$ T/K and $T_c=40.1$ K. For the $x=0.20$ sample the data give for the midpoint 1.9 T/K and 34.8 K, respectively. The values of T_c are obtained from extrapolation of the respective $0.5R_N$ and $1.0R_N$ high-field data to $B_0=0$. We also took data (not shown) using a noncontacting rf-loss technique on the $x=0.15$ sample. This measurement yields a measure of $B_{c2}(T)$ near the onset of superconductivity.⁷ Pulsed-field measurements were made in the pulsed-field facility at the laboratory up to 45 T at 4.2 K using the rf technique. For both the $x=0.20$ and $x=0.15$ sample, it was clear that a fraction of each sample was superconducting at 45 T. These data are indicated by the arrow in Fig. 2.

Comparison of the results for the two samples shows the following: (1) the higher- T_c material ($x=0.15$) has a higher $B_{c2}(T)$ and higher midpoint resistive slope $(dB_{c2}/dT)_{T=T_c}$ than the $x=0.20$ sample, (2) even in the $x=0.15$ sample, with a $B_0=0$ resistive transition width of 2 K, there is a large increase in the width of the resistive transition as T decreases. If $0.1R_N$ to $0.5R_N$ data are used as a measure of the half-width of the entire transition ($0.1R_N$ to $1.0R_N$) then this width is about 24 T at 30 K, (3) the values for $B_{c2}(T)$ at $0.5R_N$ are conservative estimates for the upper limits of $B_{c2}(T)$, and (4) values of $B_{c2}(T)$ far above those reported for any superconductor to date are obtained for the highest- T_c fractions. Whether the breadth of these transitions is largely due to inhomogeneity in the materials, or anisotropy in $B_{c2}(T)$, or both is not clear. Single-crystal measurements would be very useful to clarify this aspect.

Figure 2 shows the range of $B_{c2}(T)$ predicted by midpoint and onset data near T_c for $x=0.15$ using the standard $B_{c2}(T)$ theory for a three-dimensional dirty type-II superconductor.⁸ $B_{c2}(0)$ ranges from 50 to 140 T. [For convenience we chose the upper limit for the onset data as $(dB_{c2}/dT)_{T=T_c}=5.0$ T/K.] The upper bound assumes that the spin-orbit scattering parameter $\lambda_{SO}=\infty$, so that

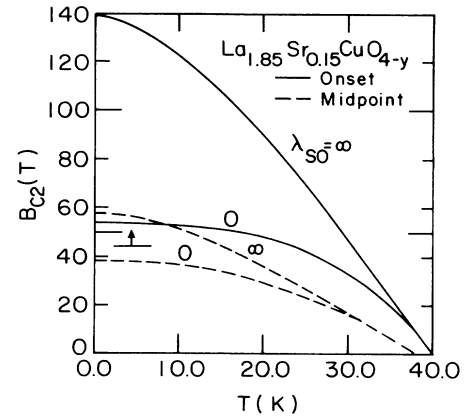


FIG. 2. Calculated upper ($\lambda_{SO}=\infty$) and lower ($\lambda_{SO}=0$) bounds for $B_{c2}(T)$ using $B_{c2}(T)$ for $0.5R_N$ (dashed curves) and $1.0R_N$ (solid curves) data of Fig. 1 for $\lambda_{e-ph}=0$. Points at 0, 4.2, and 27 K are listed in Table I for reasonable values of λ_{e-ph} . The arrow indicates that there is a fraction of the material which is superconducting above 45 T as measured with pulsed fields at 4.2 K.

the superconducting pairs are broken only by the orbital pair breaker, whereas the lower bound assumes $\lambda_{SO}=0$, so that these pairs are broken by both the orbital and Pauli-spin pair breakers. The dashed curves give the range of $B_{c2}(T)$ for the midpoint $B_{c2}(T)$ data and the solid curves are the range $B_{c2}(T)$ for the onset $B_{c2}(T)$ data. An increased $B_{c2}(T)$ from the lower bound is calculated for increased λ_{SO} or by an increased Pauli pair breaker H_P (which includes many-body renormalization by the antisymmetric Fermi-liquid parameter,^{9,10} and here is estimated as the electron-phonon coupling constant λ_{e-ph}). Table I lists values of $B_{c2}(T)$ at $T=0, 4.2,$ and 27 K (liquid Ne) for various λ_{e-ph} 's. (The upper bound $\lambda_{SO}=\infty$ is not affected by choice of λ_{e-ph} .) These values can be used to make projections for possible high-field tests. Based on our experience with conventional superconductors, we believe that the onset data reflect the upper limit expected for homogeneous material, whereas the midpoint data give a conservative limit.

Both Figs. 1 and 2 point out several difficulties for extending experiments to higher fields in these high- T_c materials. With the present relatively narrow $B_0=0$ resistive transition materials, calculations show that dc fields of 30 T may be sufficient to narrow the range of estimated $B_{c2}(0)$ somewhat.

The temperature dependence of the resistance $\rho(T)$ in the normal state (which is large here²) also affects the estimate of $B_{c2}(T)$. In the dirty limit we assume that we can multiply the orbital pair breaker by $\rho(T_c)/\rho(T)$. Inclusion of $\rho(T)$ shows that $B_{c2}(27$ K) is hardly affected, but $B_{c2}(0$ K) and $B_{c2}(4.2$ K) can be reduced by as much as $\rho(0)/\rho(T_c)$. If $\rho(T)$ decreases linearly to 20 K and then remains constant, $\rho(20)/\rho(T_c)=0.65$ for $x=0.15$.² Further reduction in $\rho(T)$ can lead to a sufficient reduction in $B_{c2}(T)$ to show a reentrantlike shape at low T . The above discussion suggests that extrapolations of $B_{c2}(T)$ to low temperatures may be open to a wide variation; direct

TABLE I. Estimates of $B_{c2}(T)$ based on high-field data near T_c .

Estimate based on onset of data; $(dB_{c2}/dT)_{T=T_c} = 5.0$ T/K, $T_c = 40.1$ K				
λ_{e-ph}	λ_{SO}	$B_{c2}(0$ K)	$B_{c2}(4.2$ K)	$B_{c2}(27$ K)
...	∞	140 T	135 T	59 T
0	0	52	50	38
1	0	84	84	51
2	0	104	104	55
Estimate based on midpoint data; $(dB_{c2}/dT)_{T=T_c} = 2.2$ T/K, $T_c = 38.1$ K				
λ_{e-ph}	λ_{SO}	$B_{c2}(0$ K)	$B_{c2}(4.2$ K)	$B_{c2}(27$ K)
...	∞	58	57	23
0	0	38	38	21
1	0	50	50	23
2	0	54	53	23

measurements are necessary and these will require very high fields.

The measured $(dB_{c2}/dT)_{T=T_c}$, T_c , and $\rho(T_c)$ yield superconducting and normal-state parameters.¹⁰ For $(dB_{c2}/dT)_{T=T_c} = 5$ T/K, $\rho(T_c) = 430 \mu\Omega \text{ cm}$ (Ref. 2), and $T_c = 40$ K, the electronic specific heat $\gamma = 2600 \text{ erg cm}^{-3} \text{ K}^{-2}$, the Ginsburg-Landau coherence length $\xi_{GL}(0)$, and penetration depth $\lambda_{GL}(0)$ are 1.3 and 210 nm, respectively. The above parameters assume that these samples are in the dirty limit with carrier densities $> 10^{22} \text{ cm}^{-3}$. Estimates based on $(dB_{c2}/dT)_{T=T_c} = 2.2$ T/K give $\gamma = 1100 \text{ erg cm}^{-3} \text{ K}^{-2}$, and comparable GL lengths. It should be noted that the value of γ , although low, is comparable to that of NbN (Ref. 11) ($\gamma = 1700 \text{ erg cm}^{-3} \text{ K}^{-2}$). Furthermore, the quoted ρ values for the high- T_c materials are probably upper limits. The estimates above indicate that the La-Sr-Cu-O system is probably a conventional BCS superconductor as suggested.⁶

The large values of $B_{c2}(T)$ projected for these new superconductors will make measurements at low temperature very difficult. To test whether paramagnetic limiting is appreciable,¹² measurements can be extended with dc or pulsed fields at liquid Ne temperatures. An experimental magnet using a new Cu/Nb microcomposite has been tested up to 68.4 T at the Francis Bitter National Magnet Laboratory¹³ and can be used for selected measurements. Such experiments are planned for the near future.

On completion of this work we received a copy of work by Capone II, Hinks, Jorgensen, and Zhang¹⁴ which reports $B_{c2}(T)$ measurements to 6 T on $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ with a midpoint $(dB_{c2}/dT)_{T=T_c} = 2.13$ T/K, midpoint $T_c = 34.7$ K, and extrapolated $B_{c2}(0) = 50$ T. (Data by Tarascon *et al.*² taken to 5 T on our samples also indicated that these materials are high-field superconductors.) Because of the broad transitions and curvature near T_c we would expect low-field data (~ 5 –6 T) to underestimate $B_{c2}(T)$. Our reported midpoint slope for the $x = 0.15$ sample is in very good agreement with the lower-field data of these authors, but our midpoint is $T_c = 38.1$ K. On the other hand, our midpoint slope for the $x = 0.20$ sample is 1.9 T/K and has a midpoint $T_c = 34.8$ K, very close to these authors' $x = 0.15$ sample. Our high-field data thus confirm these earlier projections that these high- T_c materials will have very high upper critical fields.

We wish to thank T. H. Geballe, J. M. Rowell, and J. H. Wernick for valuable discussions. We also wish to thank M. J. Neal for useful discussions. Three of us (J.M.T., K.A.D., and T.P.O.) acknowledge support of National Science Foundation Industry-University Cooperative Research program through NSF Contract No. DMR-8403493. The Francis Bitter National Magnet Laboratory is supported by the NSF.

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

²J. M. Tarascon, L. H. Greene, W. R. McKinnon, G. W. Hull, and T. H. Geballe, *Science* **235**, 1373 (1987).

³S. Uchida, K. Takagi, K. Kitazawa, and S. Tanaka, *Jpn. J. Appl. Phys. Lett.* (to be published).

⁴H. Takagi, S. Uchida, K. Kitazawa, and S. Tanaka, *Jpn. J. Appl. Phys. Lett.* (to be published).

⁵C. W. Chu, P. H. Hor, R. L. Mong, L. Gao, Z. J. Huang, and Y. O. Wang, *Phys. Rev. Lett.* **58**, 405 (1987).

⁶R. Cava, R. B. van Dover, B. Batlogg, and E. A. Rietman, *Phys. Rev. Lett.* **58**, 408 (1987).

⁷See, for example, a direct comparison of resistive and rf measurements in Nb_3Ge , S. Foner, E. J. McNiff, Jr., J. R. Gavaler, and M. A. Janocko, *Phys. Lett.* **47A**, 485 (1974).

⁸A. L. Fetter and P. C. Hohenberg, in *Superconductivity*, edited

by R. P. Parks (Marcel Dekker, New York, 1969), Vol. II, p. 866.

⁹J. A. X. Alexander, T. P. Orlando, D. Rainer, and P. M. Tedrow, *Phys. Rev. B* **31**, 5811 (1985).

¹⁰T. P. Orlando, E. J. McNiff, Jr., S. Foner, and M. R. Beasley, *Phys. Rev. B* **19**, 4545 (1979).

¹¹C. Geibel, H. Rietschel, A. Jounod, M. Pelizzone, and J. Müller, *J. Phys. F* **15**, 405 (1985).

¹²It should be noted that evidence for Pauli paramagnetic limiting was reported in one of the earlier superconducting oxide compounds Li-Ti-O, which had a $T_c = 11.2$ K and values of $(dB_{c2}/dT)_{T=T_c}$ ranging up to 3.9 T/K; S. Foner and E. J. McNiff, Jr., *Solid State Commun.* **20**, 995 (1976).

¹³S. Foner, *Appl. Phys. Lett.* **49**, 982 (1986).

¹⁴D. W. Capone II, D. G. Hinks, J. D. Jorgensen, and K. Zhang, *Appl. Phys. Lett.* (to be published).