Title
SPECIFIC HEAT OF THE HIGH T SUPERCONDUCTORS (La,M)2Cu04 AND YBa2Cu307 IN MAGNETIC FIELDS

Author
Phillips, N.E.

Publication Date
1987-08-01

LBL-23872

Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA

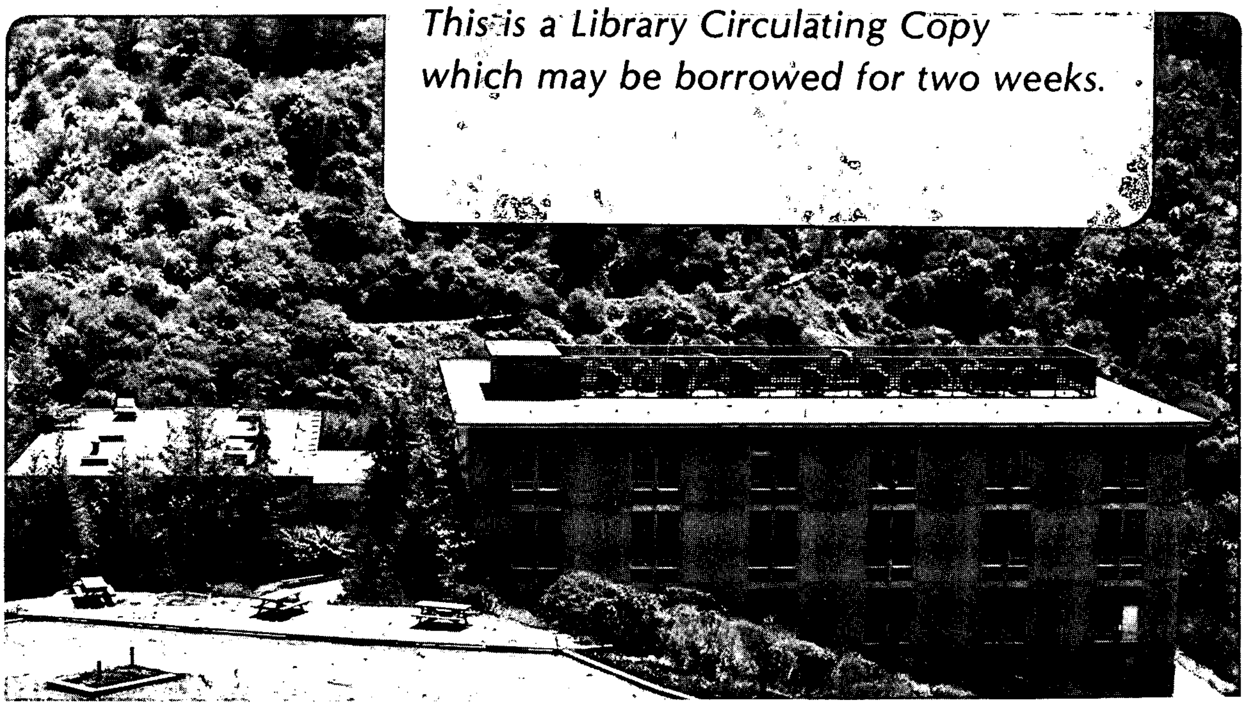
Materials & Chemical Sciences Division

Presented at the Yamada Conference XVIII
on Superconductivity in Highly Correlated Fermion Systems,
Sendai, Japan, August 31–September 3, 1987,
and to be published in Physica B

Specific Heat of the High T_c Superconductors $(La,M)_2CuO_4$ and $YBa_2Cu_3O_7$ in Magnetic Fields

N.E. Phillips, R.A. Fisher, S.E. Lacy, C. Marcenat, J.A. Olsen,
W.K. Ham, A.M. Stacy, J.E. Gordon, and M.L. Tan

August 1987



Prepared for the U.S. Department of Energy under Contract DE-AC03-76SF00098

DISCLAIMER

SPECIFIC HEAT OF THE HIGH T_c SUPERCONDUCTORS $(La,M)_2CuO_4$ AND $YBa_2Cu_3O_7$ IN MAGNETIC FIELDS*

N. E. Phillips, R. A. Fisher, S. E. Lacy, C. Marcenat, J. A. Olsen, W. K. Ham, A. M. Stacy, J. E. Gordon[+] and M. L. Tan[+]

Materials and Chemical Sciences, Lawrence Berkeley Laboratory, University of California, Berkeley, CA 94720

The specific heats, C, of the high T_c superconductors $YBa_2Cu_3O_7$, $La_{1.85}Ca_{0.15}CuO_4$ and $La_{1.85}Sr_{0.15}CuO_4$ have been measured from 0.4K to above the superconducting transition temperature in magnetic fields, H, from 0 to 7T. From measured changes in C with H, values of $\gamma(H)=[C(H)/T]_{T=0}$ and $\Delta C/T_c = [(C_s-C_n)/T]_{T=T_c}$ have been evaluated. These results have been used to estimate the normal state γ, the fraction of the sample that is superconducting in zero field, f_s, and the value of the upper critical field, $[H_{c2}]_{T=0}$.

The specific heat, C, of a 22.7 g $YBa_2Cu_3O_7$ (YBCO) sample was measured using a standard heat pulse method from 0.4 to 30K in zero magnetic field, H, and in fields of 3.5 and 7T. The specific heat was also measured between 80 and 95K by a continuous heating method (dT/dt was about 1.5mK/s) in zero field and at 7T. An estimate of the precision for both types of measurements was about 0.1%. In zero field C/T below about 1.5K has an upturn which may be intrinsic but is probably due to the presence of a magnetic impurity such as $BaCuO_2$. $BaCuO_2$ is usually present in YBCO, and furthermore, a spectroscopic analysis detected no other impurities at a level of <0.1 wt.%. In fields of 3.5 and 7T the C/T data clearly show Schottky-like anomalies, with maxima near 1.5 and 3K, respectively, which are presumably due to the same magnetic impurity causing the upturn in zero field. From the amplitude of the maxima, and assuming two electronic states, the amount of impurity is estimated to be about 0.4 mole%. (Previous measurements on another YBCO sample had an anomaly in C/T near 1K, which in all likelihood arose from a 1 mole % Cr impurity which was found in a subsequent spectroscopic determination.) Meissner effect measurements in $H = 8.62$G (calibrated using the normal/superconducting transition in a Sn sphere) indicated an onset T_c of 92K with a transition width (10-90%), ΔT_c, of 8K. The sample had a flux exclusion $-4\pi\chi_V = 0.23$, based on the total volume of the sample and corrected for demagnetizing effects, assuming the cylindrical sample (diameter equal to height) could be approximated by a sphere of the same diameter.

The low temperature specific heat data were fitted to

$$C(H) = A(H)/T^2 + \gamma(H)T + B_3T^3 + B_5T^5, \qquad (1)$$

in the range T_L-15K. T_L was selected so that the T^{-2} approximation to the Schottky anomaly was valid. (In 7T, below about 1K, there is also a T^{-2} component due to hyperfine contributions.) All coefficients in Eq. 1 were evaluated for one mole of YBCO, for which the molecular weight was taken as 666 g/mole. In zero field $\gamma(0)$ is 7.1 mJ/K^2·mole YBCO. For 3.5 and 7T, $\gamma(H)$ was, respectively, 7.8 and 8.6 mJ/K^2·mole YBCO. B_3 and B_5 were field independent to within about 1%, in contrast to the field dependence [1] of B_3 found for the $(La,M)_2CuO_4$ compounds. From B_3, a Debye temperature, θ_D, of 430K was evaluated from $B_3/13 = (12/5)\pi^4/\theta_D{}^3$. Figure 1 is a plot of $[C-C_1]/T$ vs T below 20K, which shows the anomalies due to the presumed magnetic impurity and the derived values of $\gamma(H)$ as horizontal lines. (The addenda heat capacity is not yet known to high accuracy. Final values of C for the sample will be reported in the future. The shape of the curves in Fig. 1 may be slightly altered when the corrected addenda are subtracted.)

While some current theoretical models for high T_c superconductors [2] predict a linear contribution to the specific heat in the superconducting phase, our tentative analysis of the experimental results assumes that the non-zero value of $\gamma(0)$ is evidence that in zero field a fraction of the sample remains in the normal phase down to the lowest temperatures measured. We emphasize that the experimental data do not permit a choice between these two possibilities.

The low temperature specific heat measurements on YBCO in magnetic fields of 3.5 and 7T show that the γ increases approximately linearly with magnetic field, $d\gamma(H)/dH = 0.21$ mJ/K^2 T mole YBCO. A similar dependence of γ on H has been observed in the $(La,M)_2CuO_4$ compounds (see Table I) as well as in other type II superconductors.

For zero field, the heat capacity data show the anomaly associated with the normal/superconducting transition between 88 and 92K. The anomaly is suppressed and shifted downward by the application of 7T. The shift is consistent with the estimates of $[dH_{c2}/dT]_{H=0}$ of 1-5 T/K from magnetic measurements[3]. A plot of [C(sample)+C(addenda)]/T vs T is shown in Figure 2. Since the heat capacity of the addenda has not been measured in the 80-100K region, absolute specific heat results cannot yet be obtained from these data. However, subtraction of the 7T data from the zero field data (shown in Figure 3) does permit an evaluation of an ideal $\Delta C/T_c \approx 38$ mJ/K$^2\cdot$ mole YBCO, comparable to the value of 39 mJ/K$^2\cdot$mole YBCO obtained by Junod et al [4]. If we assume that BCS theory is applicable, and that only a fraction, f_s, of the sample is superconducting, then $\Delta C/T_c = 1.43 f_s\gamma$, where γ is the coefficient of the linear contribution to the specific heat of the normal material. Since $\gamma(0) = (1-f_s)\gamma$, we can use these two relations, plus the experimental results $\Delta C/T_c = 38$ mJ/K^2·mole YBCO and $\gamma(0)=7.1$ mJ/K^2·mole YBCO, to obtain $\gamma = 34$ mJ/K^2·mole YBCO and $f_s = 0.79$. From our results, and the assumption that $\gamma(0) = \gamma-[d\gamma(H)/dH][H_{c2}]_{T=0}$, we estimate that for YBCO $[H_{c2}]_{T=0} = 130T$, a value which is in the range of other estimates [3]. (It is, of course, possible that the linear dependence of γ on H breaks down for sufficiently large applied magnetic fields. Our value for $[H_{c2}]_{T=0}$ should probably be regarded as a lower bound on the actual value.)

Table I contains a summary of the results on YBCO. For purposes of comparison, we have included our data on $La_{1.85}M_{0.15}CuO_{4-y}$, where M=Ca and Sr. These latter results have been described in detail elsewhere [1].

*Supported by the Director, Office of Energy Research, Office of Basic Energy Sciences, Materials Sciences Division of the U.S. Department of Energy under Contract DE-AC03-76SF00098. Additional support for J. E. G. and M. L. T. was provided by an EXXON Education Grant from the Research Corporation and for J. E. G. through the Associated Western Universities - Department of Energy Sabbatical Participant Program.

+Permanent address: Physics Department, Amherst College, Amherst, MA 01002.

REFERENCES

1) N. E. Phillips, R. A. Fisher, S. E. Lacy, C. Marcenat, J. A. Olsen, W. K. Ham, and A. M. Stacy, LT-18, Kyoto, Japan, Aug. 1987, to be published.

REFERENCES (continued)

2) P. W. Anderson, G. Baskaran, and T. Hsu, Phys Rev. Lett. 58 (1987), 2790.

3) T. P. Orlando, K. A. Delin, S. Foner, E. J. McNiff, Jr., J. M. Tarascon, L. H. Greene, W. R. McKinnon, and G. W. Hull, Phys. Rev. B35 (1987), 7249.

4) A. Junod, A. Bezinge, D. Cattani, J. Cors, M. DeCroux, O. Fischer, P. Genoud, L. Hoffman, J.-L. Jorda, J. Muller, and E. Walker, LT-18, Kyoto, Japan, Aug. 1987, to be published.

Table I.[a,b] Parameters characterizing the high-T_c superconductors $(La,M)_2CuO_4$ and $YBa_2Cu_3O_7$. (All units are in mJ, K and T.)

	$-4\pi\chi_V$	T_c	ΔT_c	$\gamma(0)$	$d\gamma/dH$	$B_3(0)$	dB_3/dH	B_5	θ_D	f_s	$\Delta C/T_c$	γ
Ca1	0.26	22	6	3.05	0.035	0.145	0.0019	0.0013	450	0.31	1.92	4.4
Sr2	0.35	37	8	1.54	0.109	0.168	0.0011	0.00085	430	0.82	9.9	8.6
Y2	0.23	92	8	7.1	0.21	0.32	0	0.0008	430	0.79	38	34

[a] $\Delta C/T_c$ are the ideal transition discontinuities from entropy conserving constructions of the broadened measured $\Delta C/T_c$.

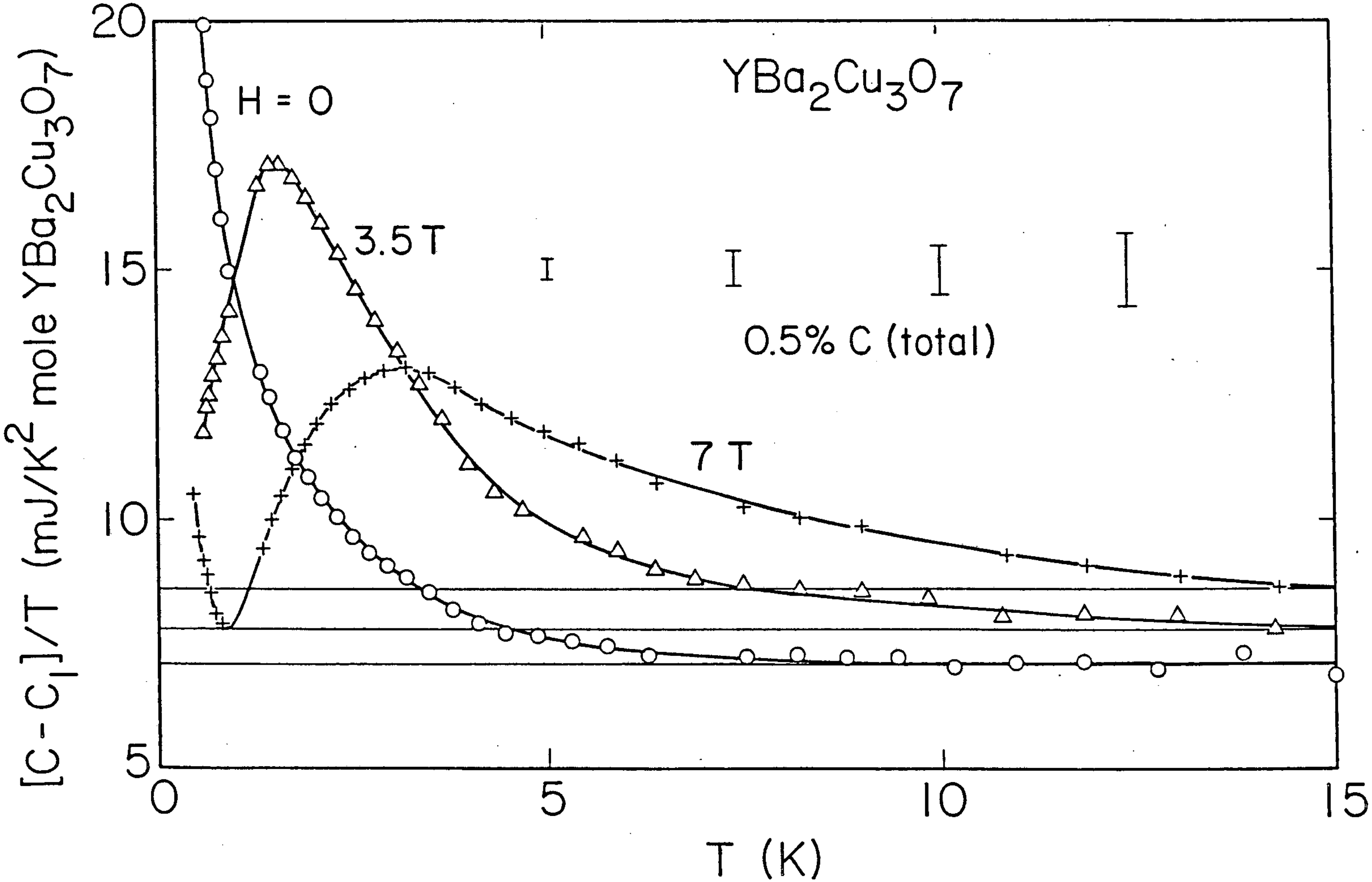
[b] γ is derived assuming $\Delta C/T_c(\text{ideal}) = 1.43\ f_s\gamma$.

FIGURE CAPTIONS

Fig. 1. $[C-C_1]/T$ vs T for $YBa_2Cu_3O_7$. The light lines are spline fits to the data, while the heavy horizontal lines represent $\gamma(H)$ at 0, 3.5 and 7T derived from fits to the data described in the text. In 7T, below about 1K, the upturn in C/T is due to hyperfine components of $^{63,65}Cu$ and ^{89}Y.

Fig. 2. [C(sample)+C(addenda)]/T vs T for $YBa_2Cu_3O_7$ at H=0 and 7T. For clarity, only one-half of the data at each field are plotted. C was measured by a continuous heating method.

Fig. 3. [C(0)-C(7T)]/T vs T for $YBa_2Cu_3O_7$ showing the onset of the normal/superconducting transition near 92K. The dashed vertical line is the idealized, entropy-conserving construction for a sharp transition at T_c.



$YBa_2Cu_3O_7$
H = 0
3.5 T
7 T
0.5% C (total)
$[C - C_I]/T$ $(mJ/K^2$ mole $YBa_2Cu_3O_7)$
T (K)
20
15
10
5
0
5
10
15

Fig. 1

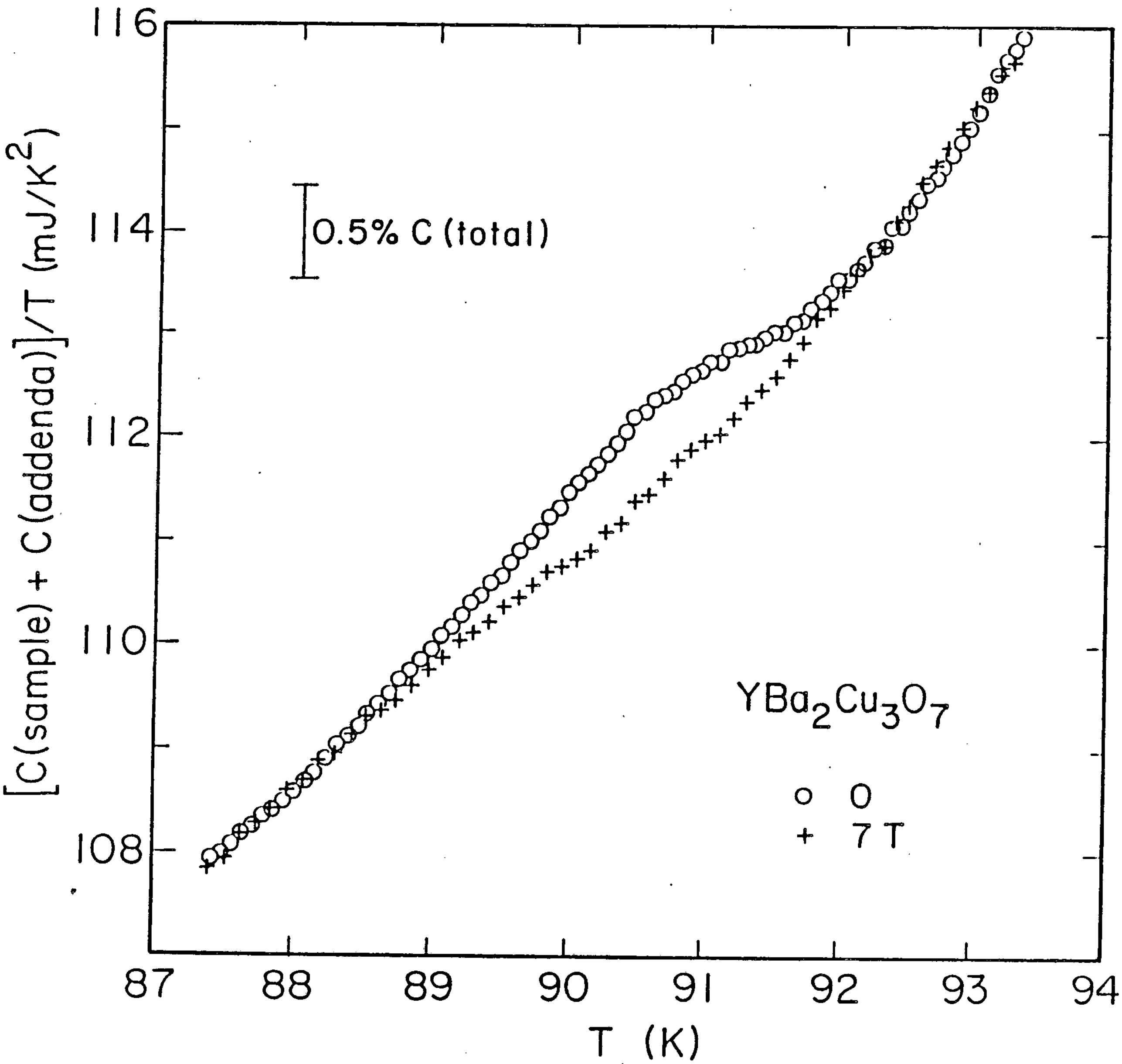

116
114
112
110
108
0.5% C (total)
[C(sample) + C(addenda)]/T (mJ/K^2)
87
88
89
90
91
92
93
94
T (K)
$YBa_2Cu_3O_7$
o 0
+ 7 T

Fig. 2

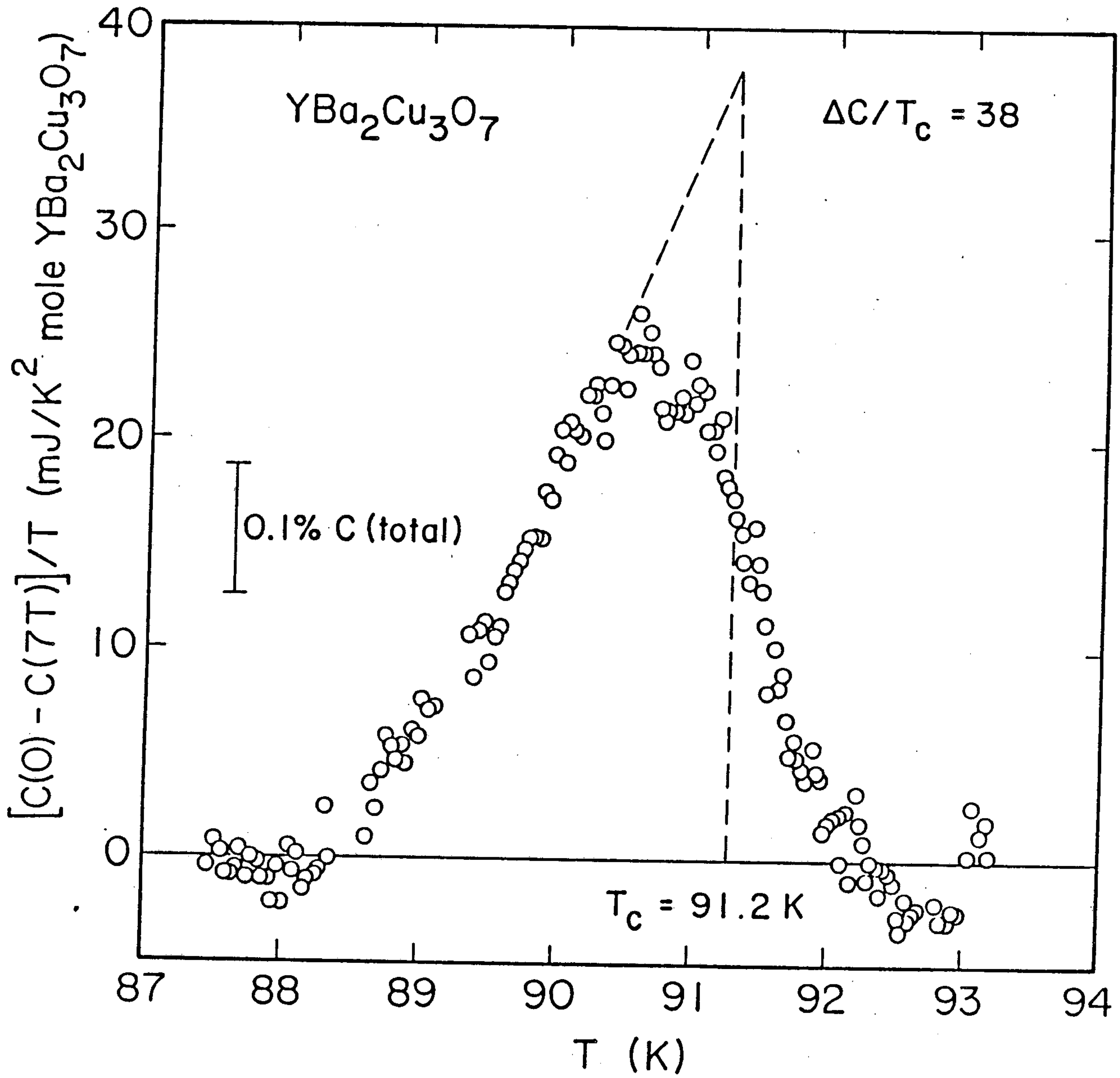

YBa2Cu3O7
ΔC/Tc = 38
0.1% C (total)
Tc = 91.2 K
[C(0) - C(7T)]/T (mJ/K2 mole YBa2Cu3O7)
T (K)
40
30
20
10
0
87
88
89
90
91
92
93
94

Fig. 3

LAWRENCE BERKELEY LABORATORY
TECHNICAL INFORMATION DEPARTMENT
UNIVERSITY OF CALIFORNIA
BERKELEY, CALIFORNIA 94720