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Publication Date

1991-09-01

LBL-31328
Preprint

Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA

Materials & Chemical
Sciences Division

Submitted to Philosophical Magazine B

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September 1991

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

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LBL-31328

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This work was 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 No. DE-AC03-76SF00098.

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ABSTRACT

Specific heat data on the high temperature superconductor $YBa_2Cu_3O_{7-\delta}$ provide evidence for strong coupling. The data on ceramic and single crystal samples also indicate that when there is a fluctuation contribution to the specific heat anomaly at T_c, the anomaly is reduced in height. These results are consistent with the the idea that an oxygen deficiency both reduces γ and enhances fluctuation effects.

1. INTRODUCTION

Shortly after the discovery of high temperature superconductors (HTSC) (Bednorz and Müller 1986), numerous measurements of the specific heat, C, of the lanthanum and yttrium oxide superconductors were reported [for extended discussions of the specific heats of HTSC's and the relevant references see Fisher et al. (1988), Junod (1990), Phillips et al. (1992)]. The specific heat data provided less dramatic evidence of superconductivity than did resistivity and magnetic measurements, but they did show a measurable anomaly at T_c (see Fig. 1 for an example of relatively recent data) and thereby demonstrated unambiguously that the superconducting transition characterized a sizeable fraction of bulk material and not merely small filamentary regions. It was recognized that the shape of this anomaly could also test the applicability of the Bardeen-Cooper-Schrieffer (BCS) theory of superconductivity. In this paper we shall discuss several aspects of this test as it applies to the case of $YBa_2Cu_3O_{7-\delta}$ (YBCO).

2. SPECIFIC HEAT MEASUREMENTS AND STRONG COUPLING

The specific heat data indicated that the new HTSC's, unlike conventional superconductors, had a contribution at low temperatures that was linear in T. Such a linear term, if intrinsic to the superconducting phase, would be inconsistent with the gap in the electron density of states characteristic of BCS theory. These results therefore suggested that the BCS electron-phonon interaction was not the mechanism principally responsible for superconductivity in the oxide materials. However, low temperature specific heat measurements on probable impurity phases [see Junod (1990) for discussion and references] suggested that at least a portion of the linear term was due to these phases. Recent work carried out in Berkeley (Phillips et al.1990) indicated that none of this linear term was intrinsic to the superconducting phase, for it was concluded that the part of the linear term not associated with impurity phases came from YBCO that remained normal even at low temperatures. In addition, by correlating the low temperature data with the height of the specific heat anomalies at T_c, the Berkeley group was able to estimate both f_s, the volume fraction of superconductivity for a given sample, and γ, the coefficient of the linear term in the normal-state specific heat. γ is important for an understanding of both normal- and superconducting-state properties, but cannot be directly measured in the case of an HTSC.

Phillips et al. (1991) estimated that $\gamma \approx 16 \pm 2$mJ/(mole K^2) and, further, that $\Delta C(T_c)/T_c \approx$ 77mJ/(mole K^2) for a fully-superconducting sample [Junod (1990) reports a similar estimate of $\Delta C(T_c)/T_c$ for the 100% superconducting transition]. Such values are clearly inconsistent with the BCS weak-coupling prediction that $\Delta C(T_c)/\gamma T_c = 1.43$. This evidence for strong coupling can be more fully tested by using the "α model", a phenomenological model that has proved useful for analyzing the specific heat anomalies of several conventional superconductors in which strong coupling effects are important (Padamsee et al. 1973). In this model, the energy gap, Δ, has a BCS-like temperature dependence, but its value at 0K, Δ_o, is an adjustable parameter. Fits to measured specific heats with this model give both $\alpha = \Delta_o/kT_c$, and γ, (or, if $f_s \neq 1$, $f_s\gamma$). In effect, the shape of the anomaly determines α, and its amplitude gives γ. The solid curve in Fig.1 is an α-model fit with $\alpha = 3.4$, $f_s\gamma = 14.5$ mJ/(mole K^2), a Gaussian distribution in T_c, and a lattice background determined from the data above 96K. This value of $f_s\gamma$ is consistent with the ~16mJ/(mole K^2) obtained by Phillips et al. (1990) and also with values for γ inferred from the temperature-independent part of the high temperature magnetic susceptibility. $\alpha=3.4$ corresponds to $2\Delta_o/kT_c=6.8$, almost double the weak-coupling value of 3.53, and a result that points to extreme strong coupling. Similar results on YBCO were obtained by Loram and Mirza (1988). Coombes and Carbotte (1989) have argued that in general the α model is unlikely to predict correctly the value of Δ_o and the temperature dependence of C below T_c. Nonetheless, it is evident from Fig. 1 that the model provides a good fit to these data.

Carbotte and co-workers [see Carbotte (1990) for a full discussion and relevant references] have discussed the question of strong coupling in the HTSC at some length. They point out that the ratio $[(dC/dT)/(\Delta C/T)]_{Tc}$ depends upon coupling strength and is, moreover, independent of any assumption regarding the value of γ. They note that in the specific heat results of the Geneva group (Junod 1990) this ratio considerably exceeds the value predicted for a weakly-coupled BCS superconductor. To be sure, an experimental determination of dC/dT depends strongly upon the imperfectly-known behavior of C_ℓ, the lattice specific heat. However, any reasonable estimate of C_ℓ suggests that strong, rather than weak, coupling characterizes YBCO. Indeed, for some samples at least, the value of the ratio actually exceeds the maximum strong-coupling value consistent with Eliashberg theory (Carbotte 1990).

A different conclusion about the strength of coupling is reached, however, by comparing the "experimental" γ,16±2mJ/(mole K^2), with γ_{bs} =13-16mJ/(mole K^2),the "bare density of states" value obtained from band structure calculations (Massida et al. 1987, Krakauer et al. 1988). From this near equality of γ and γ_{bs} we can infer that there is little enhancement of γ by the electron-phonon interaction, and therefore that YBCO could be a BCS weakly-coupled superconductor, at least if the superconductivity arises from phonon coupling.

A possible resolution of this disagreement concerning coupling strength is suggested by experimental evidence that the temperature dependence of the gap of at least one HTSC ($Bi_2Sr_2CaCu_2O_8$) is not BCS-like (Brunel et al. 1991). The value of $\Delta C(T_c)$ reflects the temperature dependence of the gap at T_c and not necessarily the value of Δ_o. Although the gap measured by Brunel et al. (1991) has $\Delta_o \approx 3.3kT_c$ (essentially the weak-coupling value), Cohen and Penn (1990) demonstrated that such a gap is consistent with a $\Delta C(T_c)/T_c$ as large as that seen in YBCO because $d[\Delta^2]/dT$ is larger at T_c than is predicted by BCS theory. However, it is reasonable to ask whether such a gap is also consistent with the behavior of C below T_c since at lower temperatures C depends upon Δ_o as well as upon $d[\Delta^2]/dT$. In order to check this, we have assumed a gap for YBCO that has a temperature dependence of the form suggested by Cohen and Penn (1990) and have calculated the specific heat assuming $2\Delta_o = 3.53kT_c$ and a value for $[d(\Delta(T)^2)/dT]_{Tc}$ large enough to produce the $\Delta C(T_c)/T_c$ obtained from an entropy-conserving construction to the data shown in Fig. 1 (see Fig. 2 and the discussion below). The results are shown as the dotted line in Fig. 1. This fit is evidently inferior to that obtained by calculating the specific heat with the α model. Thus, the specific heat data seem to require strong coupling, despite the fact that $\gamma \approx \gamma_{bs}$.

3. SPECIFIC HEAT ANOMALY: CONSEQUENCES OF OXYGEN DEFICIENCY

The data of Fig.1 are replotted in Fig 2. They were obtained by the Berkeley group from continuous heating measurements made on a five gram ceramic sample prepared by standard solid state techniques. The solid curve in Fig. 2 is a representation of the most-recently published data obtained by the Illinois group in a series of impressive measurements on small single crystals of YBCO [see Inderhees et al. (1991) for these results and for references to the earlier work]. A comparison of the ceramic and single crystal data yields both expected and unexpected results. The ~5% difference in the absolute values is a little surprising, but may arise from the fact that the small sample ac technique used in the single crystal measurements, while excellent for measuring relative values of C, is difficult to calibrate with high accuracy. The most striking, but hardly unanticipated, difference between the ceramic and the single crystal results, lies in the sharpness of the transition. The almost vertical rise in the single crystal data is clear evidence for good sample homogeneity. What is unexpected, however, is that the narrow peak in the single crystal data is not higher than the broader anomaly in the ceramic data. In fact, the "discontinuity" in the ceramic data obtained from an entropy-conserving construction (solid line in Fig. 2) shows that $\Delta C(T_c)/T_c \approx 70$ mJ/(mole K^2) whereas the height of the single crystal anomaly is only ~48 mJ/(mole K^2). On the basis of their measurements on the ceramic samples Phillips et al. (1990) argued that the different heights of the discontinuities reflected different volume fractions of superconductivity. We think it unlikely that this argument can be extended to the case of a single crystal. It is more likely that the single crystal's lower jump height is due to a lower oxygen content (higher δ in $YBa_2Cu_3O_{7-\delta}$).

Two sets of measurements on ceramic samples, (Junod et al. 1989) and (Loram et al. 1991), have shown that increasing δ from ~ 0.02 to ~ 0.15 does not markedly affect the temperature at which the anomaly is a maximum, but does considerably reduce the height. Similar results have been reported on single crystals (Wühl et al. 1991). It is our belief that very carefully-prepared ceramic samples have a smaller oxygen deficiency than do most of the single crystals, and that this difference is responsible for the smaller anomaly height in the single crystal results.

Another difference between the ceramic and single crystal results is found in the high temperature "tail" of the anomaly and in the sharp rise in the single crystal specific heat just below T_c. Both features are strong indicators of a fluctuation contribution to the specific heat - a contribution that, in zero applied magnetic field, at least, appears to result from 3-d Gaussian fluctuations (Inderhees et al. 1991). These fluctuations, like the reduced peak height, may be a consequence of the (presumably) larger oxygen deficiency in the single crystal. Resistivity measurements of oxygen-deficient ceramic YBCO samples (Cooper et al. 1991) indicate that the fluctuation contribution to the conductivity increases with δ. A similar effect is present in the specific heat (Loram et al. 1991). It is known that as δ increases, oxygen is removed from the CuO chains in YBCO and also that, for $\delta \geq 0.1$, the c-axis length increases (Rusiecki et al.1990). Cooper et al. (1991) speculate that an increasing oxygen deficiency tends to decouple the CuO_2 planes and to decrease the c-axis coherence length, ξ_c. Since the fluctuation contribution to C varies as $(\xi_a \xi_b \xi_c)$, any decrease in ξ_c enhances this contribution.

It can be argued that a comparison of the ceramic and single crystal data does not confirm the absence of a fluctuation term in the former. Such a term may be present, but is masked by a distribution in T_c that is responsible for the relatively broad anomaly and thus for a "washing out" of the fluctuation effects. However, inclusion of a fluctuation term as large as that found by Inderhees et al. (1991) actually slightly worsens the fit to the ceramic data shown in Fig. 1. While the possibility of a smaller fluctuation term cannot be excluded, the absence of a high temperature tail and of the rise just below T_c in most of the ceramic data of the Geneva group (Junod 1990), lead to the conclusion that fluctuation effects are small in the majority of ceramic samples, presumably because they are more fully oxygenated than are most of the single crystals. Further support for the δ-dependence of fluctuation effects can be found in the recent specific heat data on a single crystal prepared by the grain-growth method (Bonjour et al. 1991). This sample, which was annealed under 7 bars of oxygen and was reported to be very nearly fully-oxygenated ($\delta \sim 0.03$), showed no evidence of a fluctuation contribution to C.

Fluctuation effects have been observed in a few ceramic samples. Despite the breadth of the ceramic transitions, the evidence of a fluctuation contribution is almost as striking in these cases as in that of the single crystal. However, in these cases the anomaly height is lower than that in the ceramic data reported here. Some of the reduction may be associated with a reduced fraction of volume superconductivity (Phillips et al. 1990), but it is our guess that had the oxygen content of these samples been determined, δ would have been found to be greater than the typical value for well-oxygenated ceramic samples.

It appears, then, that a careful determination of the oxygen content of YBCO samples is as essential to an understanding of the specific heat anomaly as it is to other sample characteristics. It has been shown (Farneth et al. 1989) that χ_0, the temperature-independent part of χ, decreases as δ increases, a result that indicates a similar decrease in $N(E_f)$, the electron density of states at the Fermi energy. Such a dependence upon oxygen deficiency is consistent with the calculations of Calandra and Minerva (1990). For a surer understanding of the role of δ, it would be helpful to repeat the measurements of Farneth et al. (1989) on samples for which the specific heat variation with δ had also been determined. Similarly, a careful examination of the temperature-dependent part of χ might reveal whether or not the fluctuation contribution to $\chi(T)$ reported by Lee et al. (1989) is a function of δ, as would be expected on the basis of the above arguments.

4. SUMMARY

The specific heat data near T_c support the idea that strong-coupling is present in YBCO. On the other hand, the result $\gamma \approx \gamma_{bs}$ may indicate that electron-phonon enhancement effects are small. This apparent contradiction may be a clue that the mechanism principally responsible for superconductivity in YBCO, and possibly in other high-temperature superconductors, is not the conventional electron-phonon interaction. The specific heat data on single crystals and on some ceramic samples also show fluctuation effects, but in such cases the height of the specific heat anomaly is reduced. Both effects are likely to be a consequence of oxygen deficiency. If it proves to be correct that enhanced fluctuation effects are associated with decreasing c-axis correlation lengths, then anisotropy in YBCO should increase with δ - a result that can be tested by a number of different measurements on single crystals.

ACKNOWLEDGMENTS

This work was 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-ACO3-76SF000098. Additional support for J.E.G. was provided by the Research Corporation and by Associated Western Universities, Inc.

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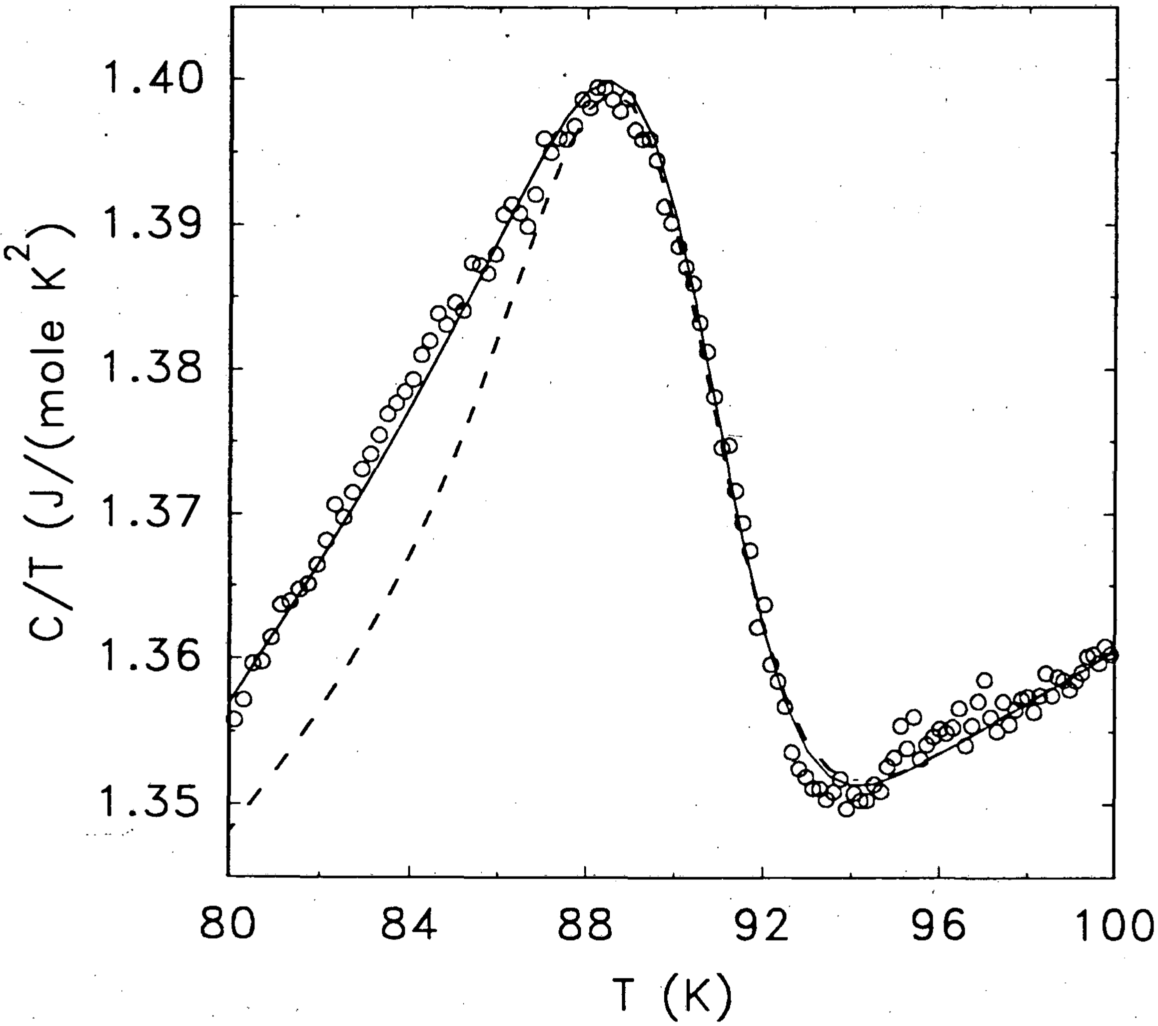
FIGURE CAPTIONS

FIGURE 1. C/T vs. T for a ceramic sample of YBCO. The solid curve is based upon the "α model" (Padamsee, et al. 1973), while the dashed curve is calculated assuming a gap similar to that obtained from reflectivity measurements on $Bi_2Sr_2CaCu_2O_8$ (Brunel et al. 1991). Both curves assume a Gaussian distribution in T_c and a lattice heat capacity obtained from the data above 96K.

FIGURE 2. The circles are the data of Fig.1 plotted on a different scale. The solid line is an entropy-conserving construction derived from these data and gives a discontinuity $\Delta C(T_c)/T_c \approx 70$ mJ/(mole K^2). The curve is a representation of single crystal data (Inderhees et al. 1991) and has an anomaly height of ~ 48 mJ/(mole K^2).



XBL 919-1983

Fig. 1

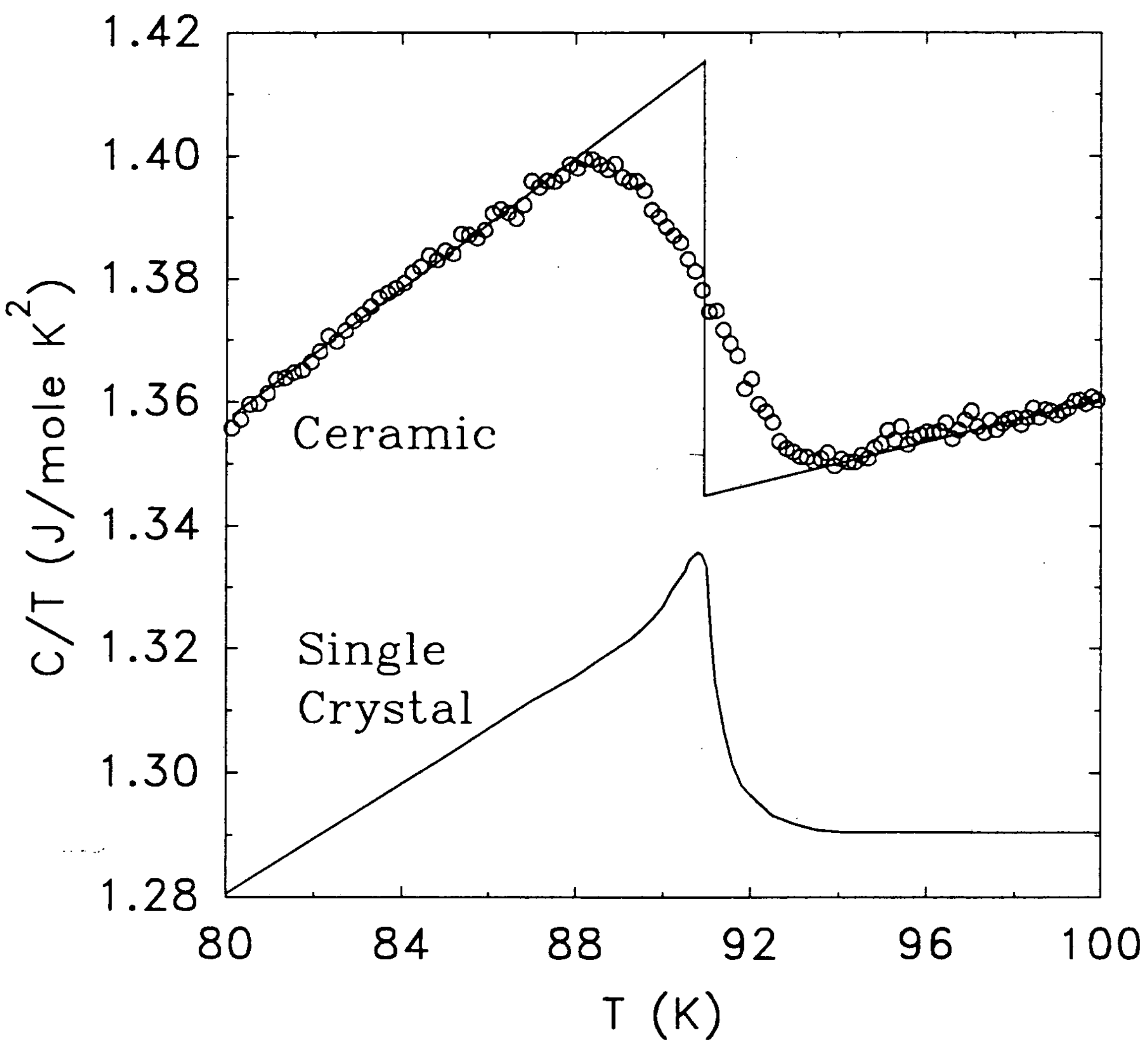


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Fig. 2

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