

## Effect of magnetic fields on the specific heat of a $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ single crystal near $T_c$

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In a  $c$ -axis magnetic field, the specific-heat peak of a single crystal of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  broadens and decreases in amplitude with little shift in position. The difference between the zero-field specific heat and that measured in a field is found to satisfy a scaling relation which suggests critical, as opposed to Gaussian, fluctuations. This is interpreted in terms of the broadening of the critical regime by the magnetic field, and suggests either renormalization of the critical exponents by disorder or an order parameter with more than two components.

Recently, we observed<sup>1</sup> the effect of fluctuations near  $T_c$  on the specific heat of single crystals of the high-temperature superconductor  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (Y-Ba-Cu-O). In addition to the (BCS) Bardeen-Cooper-Schrieffer-like jump, a three-dimensional (3D) Gaussian-fluctuation contribution was required to fit the specific-heat data. In this and other<sup>2</sup> regards, Y-Ba-Cu-O behaves, in the absence of a magnetic field, very much as expected for a strongly type-II material, but one with a very short ( $\leq 10 \text{ \AA}$ ) zero-temperature Ginzburg-Landau coherence length  $\xi(0)$ . This length plays a crucial role<sup>3</sup> in determining the temperature range over which mean-field theory approximates the behavior near  $T_c$ ; an extremely small value of  $\xi(0)$  suggests that *critical* effects must be considered.

One way to explore the critical behavior is to examine the scaling of some property as a function of two thermodynamic variables or "fields." In this Rapid Communication, we study the scaling behavior of the specific heat<sup>4</sup> as a function of temperature and magnetic field; the Illinois crystal of Ref. 1 is used. In ordinary superconductors,<sup>5</sup> the effect of the field is to shift the specific-heat step to a lower temperature  $T_c(H)$ , with little change in shape.<sup>6</sup> However, in Y-Ba-Cu-O crystals, the resistive transition is strongly broadened in applied fields<sup>7</sup> making it difficult to define  $T_c(H)$ . We find the specific-heat peak to be broadened and decreased in amplitude when a field is applied along the  $c$  axis, but with little shift in temperature.

The field dependence of the excess specific heat  $C_H$  due to Gaussian fluctuations was calculated by Lee and Shenoy;<sup>6</sup> in both clean and dirty limits it can be expressed in scaling-law form as

$$C_H(T) = h^{-1/2} f(t_H/h), \quad (1)$$

where  $h = \xi^2(0)H/\phi_0$ ,  $t_H = T/T_c(H) - 1$ , and  $\phi_0$  is the flux quantum. For large values of its argument,  $f(x) \rightarrow x^{-1/2}$ , leading to the usual<sup>8</sup> zero-field fluctuation contribution to the specific heat for  $d=3$ . As was shown previously,<sup>1</sup> our single-crystal data are consistent with this form at vanishing fields. However, as we discuss below, the magnetic-field dependence of the specific heat does *not* agree with Eq. (1), but can be represented by a scaling form similar to Eq. (1).

The single crystal used in this work was prepared using a technique similar to that of Schneemeyer *et al.*<sup>9</sup> Details will be published separately. The specific heat  $C_H$  was measured with the ac method.<sup>1,10</sup> A pair of Chromel-Constantan (type E) thermocouples was formed by crossing and spot-welding 25- $\mu\text{m}$  wires which were then attached to a (001) face of the sample with a minute amount of GE 7031 varnish. The opposite surface was darkened with colloidal graphite to enhance light absorption. Quartz fibers were glued to the thermocouple wires close to the sample to prevent sample rotation, and the entire assembly was cemented to a Mylar frame. The sample assembly was sealed in a cryostat containing He gas as the thermal link. One arm of the thermocouple detected the ac temperature oscillations induced by chopped-light heating; the second arm monitored the slight dc temperature offset of the sample from the thermal bath. The temperature was measured with carbon-glass sensor, and the usual<sup>11</sup> corrections for magnetic-field effects were made.

In Fig. 1, we show the ratio  $C_0/T$  for our sample in the vicinity of the transition temperature. This representation

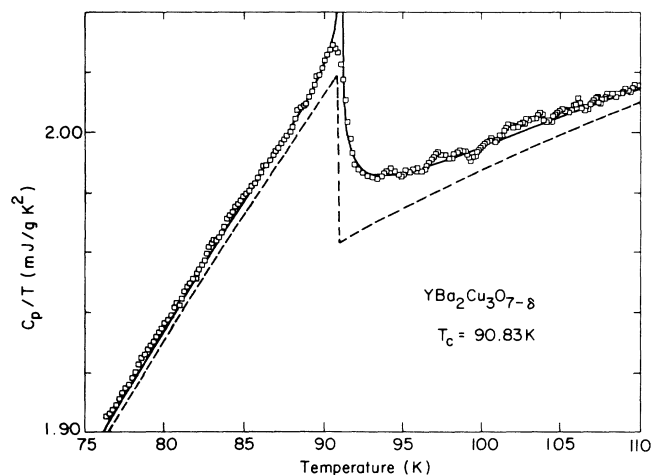


FIG. 1. Specific heat of our sample in zero applied field. The dashed line is the sum of the background and BCS-like contribution. The curvature results from plotting  $C_H/T$ . The solid curve includes Gaussian fluctuations as in Ref. 1.

reduces somewhat the strong underlying temperature variation due to the lattice background. The amplitude of the anomaly is identical to that reported earlier<sup>10</sup> for our best polycrystalline samples, suggesting strongly that this crystal is completely superconducting below  $T_c$ . We have assumed a linear background and a BCS-like step for  $C_0(T)$ , shown as a dashed curve; the solid curve includes Gaussian fluctuations.<sup>1</sup> The magnetic field was applied parallel to the  $c$  axis at temperatures above 100 K and the sample cooled to approximately 75 K in the field. This procedure guaranteed that the field in the sample was initially homogeneous; in fact, there is almost no flux expulsion from single-crystal samples for fields above 0.1 T.<sup>12</sup> The specific-heat data were collected as the sample was warmed. Since no field dependence is detected at 77 K, we normalize each run to the bulk specific heat of polycrystalline material measured at that temperature. Normalization of the data at a point above  $T_c$  does not change the results. Due to very large lattice contribution, we cannot detect small changes in the specific heat with applied field outside the transition region, and consequently cannot account for entropy shifted out of the transition region. The inset of Fig. 2 shows a representative set of data taken at 1.5 and 4.5 T, along with the zero-field data of Fig. 1. Note that there are two "branch points" at which  $C_H$  deviates from  $C_0$ . The behavior resembles that of a ferromagnet,<sup>4</sup> and there is no obvious feature that shifts downward in temperature at the rate 2 K/T, as suggested from susceptibility data<sup>7</sup>—except the low-temperature branch point. To explore the field dependence and to eliminate the lattice background, we subtract the data in a field from those taken in zero field at the same temperature, and plot the differences in Fig. 2. Both the peak height and width increase with field.

Lee and Shenoy<sup>6</sup> demonstrated theoretically that an important effect of the magnetic field is to broaden significantly the width of the critical region. To explore

this, we perform a scaling analysis based on the assumption that the magnetic field enters into the singular part of the free energy through the term  $(\mathbf{p} - 2e\mathbf{A}/c)$ ;  $\mathbf{A}$  scales, therefore, as an inverse length. As a result, the fluctuation contribution to the free energy will have the scaling form

$$F_{fl} = h^{d/2} g(t_H/h^{1/2\nu}), \quad (2)$$

where  $\nu$  is the exponent governing the divergence of the correlation length.

The specific heat associated with the fluctuation free energy (2) is of the form

$$C_H = h^{(d/2-1/\nu)} g''(t_H/h^{1/2\nu}). \quad (3)$$

Clearly, the Gaussian-model result  $\nu = \frac{1}{2}$  leads immediately back to (1), as it must. So long as we choose  $t_H = t_0 = t$  (i.e., points equidistant from the critical point) it is possible to write

$$(C_0 - C_H) h^{(1/\nu-d/2)} = f(t/h^{1/2\nu}), \quad (4)$$

where  $f(x)$  is a scaling function. Only the portion of the specific heat associated with the superconducting transition satisfies (4). Surprisingly, the curves can be scaled by choosing  $t_H$  such that the peaks align. These shift by only 0.5 K over the experimental field range, i.e.,  $t_H$  varies by  $\sim 5 \times 10^{-3}$  which is negligible. We are implicitly assuming that the true  $T_c$  for scaling purposes remains close to 91 K. Note in Fig. 2 that the peak of  $C_0 - C_H$  increases with field and requires a negative power of  $h$  on the left-hand side of Eq. (4); i.e.,  $\nu > 2/d$ . Indeed, the data can be collapsed onto a single curve by choosing a value  $\nu = 0.75 \pm 0.03$  as shown in Fig. 3. Note that the  $d=3$  Gaussian value  $\nu = \frac{1}{2}$  gives the wrong qualitative behavior while both  $d=3$  critical behavior<sup>13</sup> ( $\nu = \frac{2}{3}$ ) and  $d=2$  Gaussian fluctuations ( $\nu=1$ ) predict no field dependence for  $C_0 - C_H$  at  $t=0$ . We have noticed a tendency for the scaling behavior to break down near the peak for  $H \leq 1$  T;

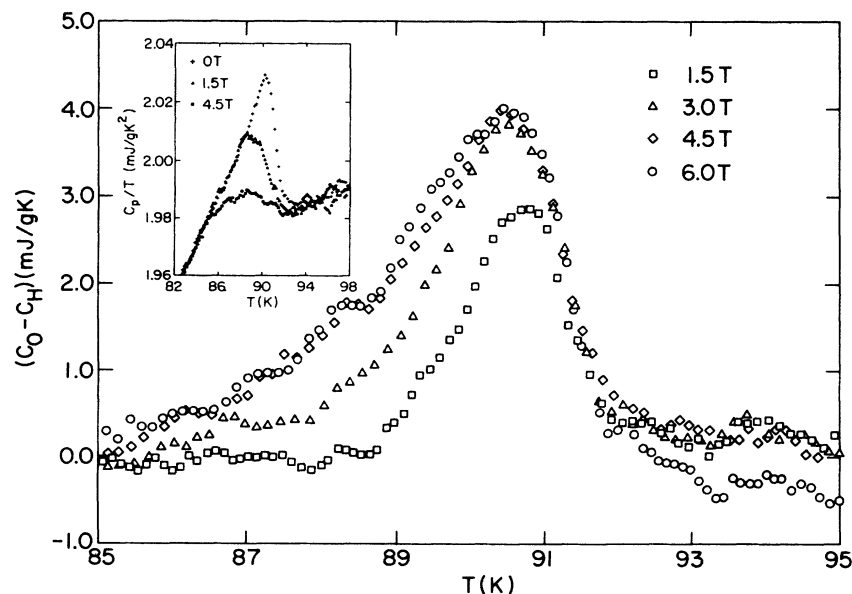


FIG. 2.  $C_0(T) - C_H(T)$  vs temperature  $T$  at various fields. Inset: Zero field, 1.5-, and 4.5-T data from which the difference data were extracted.

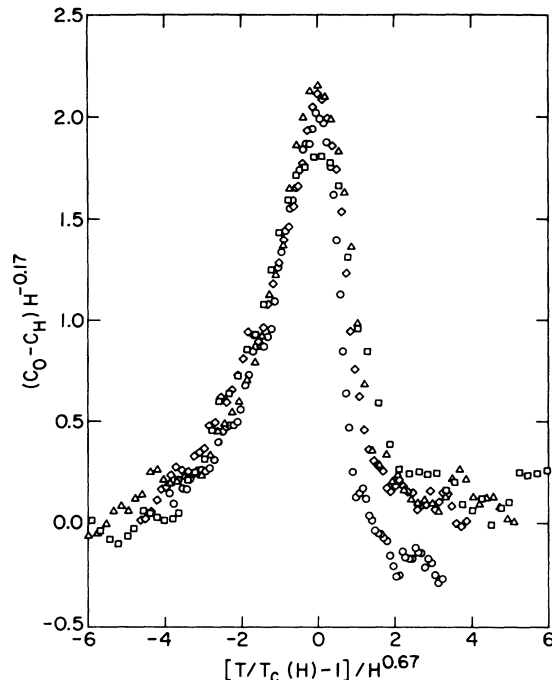


FIG. 3. Scaling of the data of Fig. 2. The values of  $T_c(H)$  used are  $T_c(1.5 \text{ T})=90.7 \text{ K}$ ;  $T_c(3.0 \text{ T})=90.5 \text{ K}$ ;  $T_c(4.5 \text{ T})=90.4 \text{ K}$ ; and  $T_c(6.0 \text{ T})=90.4 \text{ K}$ . The units of the abscissa are  $10^{-3} \text{ (kOe)}^{0.67}$ .

this may reflect rounding effects.

For the argument leading to Eq. (4) to be valid,  $C_0(T)$  must exhibit critical behavior at small  $|t|$ , with an exponent  $\alpha$  that satisfies the scaling relation  $\alpha = 2 - d\nu$ . Assuming  $d=3$ , this gives  $\alpha = -0.25 \pm 0.06$ . In Fig. 4, we plot  $\log[C_{\text{reg}} - C_0(T)]$  vs  $\log(\pm t)$ , which presumes a cusp and no discontinuity at  $T_c$ ; the plus and minus signs refer to above and below  $T_c(0)$ , respectively. In both this fit and the Gaussian fit of Fig. 1, we have modeled the regular contributions as  $C_{\text{reg}} = B + Ct$ . The solid lines in Fig. 4 correspond to  $\alpha = -0.25 \pm 0.05$  and the coefficients  $A^\pm$  of the power law are in the ratio  $A^+/A^- = 2.2 \pm 0.5$ . While no BCS step discontinuity was included in the fit, the cusped fitting function has a built-in difference of  $(A^+ - A^-) = 6.5 \text{ J/molK}$  between high- and low-temperature limits; the BCS step in Fig. 1 is  $4 \text{ J/molK}$ . The power-law fit has an rms error 50% larger than the Gaussian fit of Fig. 1 but involves two fewer parameters. We cannot rule out either fit on statistical grounds.

From Fig. 4, we note that  $C_0 - C_H$  vanishes for  $T \cong (H/272T)^{0.67}$ . Approximating this branch point instead as a linear function would give  $t \cong (H/70T)$  or  $0.77T/K$ , which compares with other estimates. It appears then that the branch point is associated with  $H_{c2}(T)$ .

A possible explanation for the effects we have observed is that the upper critical field  $H_{c2}$  depends much more strongly on composition and disorder than does the zero-

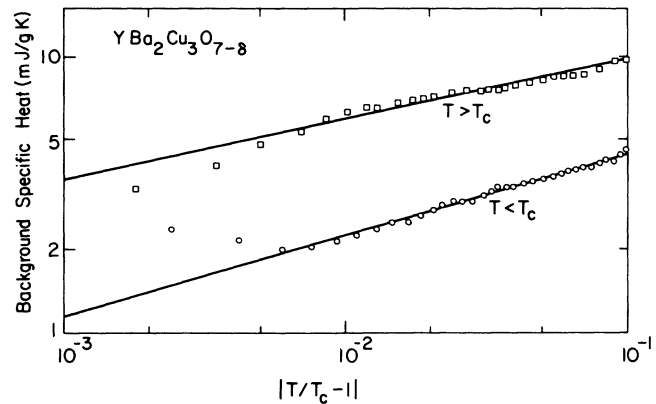


FIG. 4. Log-log plot of the difference between a regular background and the measured specific heat, assuming a cusp at  $T_c$ . The solid lines correspond to  $\alpha = -0.25 \pm 0.05$  with an amplitude ratio of  $2.2 \pm 0.5$ .

field critical temperature  $T_c$ . In this picture, the broadening simply reflects the wide range of transition temperatures induced by the applied field, rather than intrinsic behavior. However, this in itself is a deviation from the Ginzburg-Landau picture<sup>14</sup> in which  $H_{c2}$  is proportional to  $T_c$ . Demagnetizing effects are also a possible source of field inhomogeneity, but the suppression of the Meissner effect observed in large fields indicates that such effects will be minimal.

If, on the other hand, the field scaling reveals the critical behavior of Y-Ba-Cu-O, there are two possibilities for the large value of  $\nu$  and negative  $\alpha$ : renormalization of a positive  $\alpha$  due to disorder<sup>15</sup> or a large number of degrees of freedom for the order parameter. Using the epsilon expansion,<sup>16</sup> we can calculate that  $n \cong 6$  for  $\alpha = -0.25$  and  $\nu = 0.75$ . With such a large  $n$ , the  $1/n$  expansion results may be more meaningful. The  $d=3$   $1/n$  expansion gives  $n \cong 4$ . An epsilon expansion calculation for  $n=6$  and  $d=3$  predicts an amplitude ratio  $A^+/A^- \cong 2.5$  to first order in  $\epsilon$ , in good agreement with our value of  $2.2 \pm 0.5$ . This ratio is difficult to calculate accurately for the  $d=3$   $1/n$  expansion, since the spherical model predicts  $A^- = 0$  for  $d > 3$ .

The possibility of  $p$ -wave or  $d$ -wave pairing in Y-Ba-Cu-O has been suggested recently;<sup>17</sup> either could lead to  $n > 2$ . The results presented here demonstrate profound differences between the effect of a magnetic field on the superconductivity of Y-Ba-Cu-O and that of other type-II superconductors. While not conclusive, these differences may signify the existence of a more intricate pairing in Y-Ba-Cu-O than the  $s$ -wave Cooper pairs of the BCS theory.

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