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# Scaling behavior of the specific heat of a $LuBa_2Cu_3O_{7-y}$ single crystal near the $H_{c2}$ line

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High-resolution specific-heat measurements have been performed on an untwinned single crystal of $LuBa_2Cu_3O_{7-y}$ in an applied magnetic field, along the $c$ axis, of up to 5 T. The zero-field data near the normal-superconducting transition temperature ($T_c = 92.5$ K) display a very large and sharp specific-heat anomaly. Among various models for scaling the magnetic field and temperature-dependent specific-heat data, a model which incorporates the two-dimensional nature of this transition provides the best description of our data. A narrow region of crossover to three-dimensional behavior is also observed.

The intense interest in high-$T_c$ superconductors (HTSC's) originated in the confirmation of the existence of superconductivity in the Ba-La-Cu-O system near 30 K (Ref. 1) and the later discovery of superconductivity in the Y-Ba-Cu-O system near 90 K.[2] These materials exhibit two characteristics that become particularly important near the normal-superconducting transition. First, the relatively large separation between the CuO planes responsible for the superconducting behavior indicates that the two-dimensional nature of this system cannot be ignored. The coherence length at $T = 0$ K is highly anisotropic ($\xi_a \approx \xi_b > \xi_c$) and is much smaller than that of conventional superconducting materials (approximately 10 Å rather than 50 to a few thousand Å). Here $\xi_a$ and $\xi_b$ are the zero-temperature coherence lengths in the CuO plane and $\xi_c$ that in the direction perpendicular to the plane. Secondly, by virtue of the Ginzburg criterion, the very small coherence length ($\xi_0$) (Ref. 3) of the HTSC materials may make the critical fluctuation region [which is proportional to $\xi_0^{-6}$ for three-dimensional (3D) fluctuations] experimentally accessible. In principle, the temperature dependence of the critical fluctuations enables one to obtain the dimensionality of the order parameter which characterizes the nature of the normal-superconducting transition. However, analyses of the temperature dependence alone have led to no definitive conclusions.

Among many experimental probes (both microscopic and macroscopic), the specific heat is a true measure of bulk properties. High-resolution calorimetric studies have been employed by various research groups to investigate the nature of critical fluctuations.[4–8] For example, results from untwinned single crystals[6] and polycrystalline samples[7] yield noticeably pronounced specific-heat anomalies in the immediate vicinity of the transition temperature ($T_c$). These anomalies have been described by Gaussian fluctuations[6–8] and asymptotic crossover to the critical region.[9,10] A mean-field expression also gives satisfactory fittings to the specific-heat data obtained from several twinned $YBa_2Cu_3O_{7-y}$ crystals.[11] Unfortunately the large number of fitting parameters makes it difficult to unambiguously analyze the data. Separating the contributions due to critical fluctuations from those due to sample inhomogeneities and the mean-field jump in the experimental specific-heat data further complicates the procedure.

In this paper we address the situation by choosing samples with sufficiently large specific-heat anomalies and small transition widths, $\Delta T_w$,[12] so that the effects due to sample inhomogeneities are minimized,[13] and by introducing an additional experimental parameter, an applied magnetic field. We seek a consistent description of the nature of the normal-superconducting transition in the 1:2:3 compound. With this in mind, we have carried out numerous experimental runs to characterize the quality of more than one dozen high-$T_c$ superconducting single crystals. Among them, an untwinned $LuBa_2Cu_3O_{7-y}$ single crystal (hereafter denoted as Lu No. 1) exhibited a specific-heat anomaly with the largest magnitude and a small value of $\Delta T_w$. Our analysis here will be concentrated on the data obtained from this sample.

The $LuBa_2Cu_3O_{7-y}$ single crystals were grown by a self-flux technique from gold crucibles.[14] The Lu No. 1 sample was detwinned by annealing under uniaxial stress.[15] No twin boundaries are observed under reflecting polarized optical microscopy. The small single crystal (1.0 mm×0.5 mm×0.07 mm) was mounted on a thermocouple junction formed from flattened 13-$\mu$m-diam chromel and constantan wires with a minute amount of GE varnish. To maintain the sample orientation, the thermocouple wires were attached to a fiberglass ring and two thin optical fibers secured each end of the sample. The other side of the sample (irradiated with the chopped IR beam used as the ac heat source) was coated with aquadag to enhance the radiation absorption and eliminate any possible change in absorption coefficient through the transition. Any unwanted heating from the IR beam was minimized by placing a fiberglass plate with

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a hole about the size of the sample in front of the sample as a mask.

We employed an ac calorimetric system[11] to obtain the specific heat as a function of temperature and applied magnetic field of up to 5 T. The specific-heat data from the Lu No. 1 sample measured under various applied magnetic fields are shown in Fig. 1 over a 20 K temperature window. From this data, the transition width could also be determined ($\Delta T_w = 320$ mK) and the magnitude of the specific-heat anomaly was found to be $\Delta C_J = 4.5$ J/mole K.[13] Judging from these two parameters, our Lu No. 1 sample is among the best high-$T_c$ superconducting samples reported in the literature. In applied magnetic fields of up to 5 T, the onset temperature of the specific-heat anomaly on the high-temperature side does not change. However, both the peak position and magnitude of the specific-heat anomaly decrease as the magnetic field strength increases. Comparing with similar data on $YBa_2Cu_3O_{7-y}$,[16] one immediately notices that for any given magnetic field, our Lu No. 1 sample experiences a much larger reduction in the magnitude of the specific-heat anomaly than the $YBa_2Cu_3O_{7-y}$ samples does. We do not have any explanation for the difference, except that our sample may be higher in quality.[17]

In addition to a sharp drop in the specific-heat anomaly on the high-temperature side, the zero-field data exhibit a positive curvature (concave upward) just below the peak which is reproducible among different experimental runs and different thermocouple mountings. This is generally believed to signal the onset of fluctuations over the mean-field contribution.

The Gaussian fluctuation model[18] has been extensively used with a good degree of success in analyzing specific-heat data in HTSC's in zero magnetic field. In this approach, the measured specific heat is expressed as a sum of three terms: a background term ($C_b$) due to the phonon and normal electron contributions, the mean-field term ($C_{\mathrm{MF}}$), and the term due to fluctuations ($C_f$). These terms can be written as

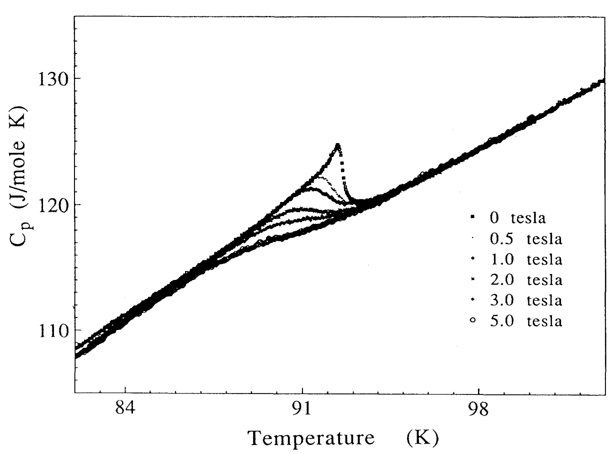


FIG. 1. Temperature dependence of specific heat of the $LuBa_2Cu_3O_{7-y}$ single crystal over a wide temperature range and with applied magnetic field up to 5 T along the $c$ axis.

$$C_b = B_0 + B_1 T \ ,$$

$$C_{\mathrm{MF}} = \gamma T(1+bt), \quad \text{for } T < T_c \ ,$$

$$C_{\mathrm{MF}} = 0, \quad \text{for } T > T_c \ ,$$

and (1)

$$C_f = A^{\pm}(T/T_c)^2 |t|^{-(2-d/2)}$$

with $A^+ = (nk_B/2)f(d)/(2\pi\xi_0)^d$, and $A^- = (k_B/2)f(d)2^{d/2}(2\pi\xi_0)^d$. The reduced temperature $t = T/T_c - 1$. The function $f(d) = \pi^2$ and $\pi$ for $d=3$ and 2, respectively. The linear dependence of the specific heat (see Fig. 1) in the high-temperature region ($96 < T < 101$ K) leads us to choose only a constant and a linear term to describe the background ($C_b$). The addition of a quadratic term ($B_2 T^2$) does not appreciably change our results. For the mean-field theory in an $O(n)$ model with quadratic fluctuations,[18] the amplitude ratio of the fluctuation term is related to $n$ (the number of components of the order parameter) and $d$ (the spatial dimension) through the relation, $A^+/A^- = n/2^{d/2}$. This approach leads to seven adjustable parameters.

Because of the layered structure of the CuO planes in $LuBa_2Cu_3O_{7-y}$,[19] we have fit our zero-field specific-heat data not only to the $d=3$ Gaussian fluctuation model but also to the $d=2$ Gaussian fluctuation model. The fitting results are satisfactory and relatively insensitive to the fitting range. Table I summarizes the fitting results over a 20 K temperature window, i.e., $81 < T < 101$ K, for $d=2$ and 3, respectively. If one compares the values of $\chi^2$ and $\xi_0$, then the $d=3$ gives a better fitting result than the $d=2$ case. A visual inspection of the fittings is less conclusive (Fig. 2). The only substantial deviation from the data for either model is within 0.5 K of $T_c$. The 3D fitting gives a fairly reasonable value for the zero-temperature coherence length ($\xi_0 \approx 8.6$ Å). The value of $\xi_0$ for the 2D fitting is rather large. The mean-field coefficient $b$ is roughly twice that for the BCS weak-coupling limit ($b = 1.72$) but within the range expected for strong-coupling superconductors. Based on the standard data analysis, the best-fitting result yields $n = 3.6 \pm 0.4$, which suggests that the fluctuation order parameter is not simply $XY$-like ($n=2$). However, the previously mentioned seven adjustable parameters may be too many to make any justifiable conclusions about the dimensionality of the system or the symmetry of the order parameter based solely on the values of $\chi^2$ and $\xi_0$. If we fix $n=2$, for example, and let the other six parameters vary, the standard deviation $\chi^2$ only increases by 20% with respect to $\chi^2_{\mathrm{min}}$ for $n=3.6$. Overall, we believe that the zero-field data alone are not sufficient to conclusively determine the best model.[20]

Further insight into the nature of the transition can be gained by obtaining and analyzing data as a function of applied field, as well as temperature. First, we perform a scaling analysis. This is a standard, powerful approach to gain physical information from experimental data (specific heat in our case) obtained as a function of two thermodynamic parameters. We investigate the scaling behavior of our data in Fig. 1 according to descriptions

TABLE I. Fitting results of zero-field data. Models I and II are the mean-field models with 3D and 2D Gaussian fluctuations, respectively. $\Delta T$ indicates the small window above and below $T_c$ within which the data are excluded from our fitting.

| | Fitting range (K) | $T_c$ (K) | $\Delta T$ (K) | $B_0$ (J/mole K) | $B_1$ (J/mole K$^2$) | $\gamma\times10^2$ (J/mole K$^2$) | $b$ | $\xi_0$ (Å) | $n$ | $\chi^2\times10^3$ |
|---|---|---|---|---|---|---|---|---|---|---|
| I | 81–101 | 92.53 | 0.07– −0.21 | 14.32 | 1.12 | 5.12 | 3.82 | 8.6 | 3.6 | 4.70 |
| II | 81–101 | 92.42 | 0.15– −0.26 | 18.61 | 1.08 | 4.98 | 5.81 | 46.0 | 4.0 | 6.88 |

based on both $d=3$ and $d=2$ fluctuations in a magnetic field.[21] In general the scaling relation is

$$\Delta C/C_{\mathrm{MF}}=g_d([T-T_c(H)]/\Delta T_d)\ , \qquad (2)$$

where $\Delta C=C-C_b$, the function $g_d$ is the scaling function for dimensionality $d$, and $T_c(H)$ is the transition temperature at the given applied magnetic field $H$, as determined by the inflection point of the high-temperature side of the specific-heat anomaly. One has $\Delta T_3=T[k_BH/(4C_{\mathrm{MF}}\phi\xi_c)]^{2/3}$ for $d=3$ and $\Delta T_2=T[2k_BH/(C_{\mathrm{MF}}\phi\delta)]^{1/2}$ for $d=2$.[22] Here $\delta\approx11$ Å is the interlayer distance, $k_B$ is the Boltzmann constant, and $\phi$ is the superconducting flux quantum.[23] We take $\xi_c=3$ Å. The results are shown in Fig. 3(a) and 3(b) for $d=3$ and $d=2$, respectively. The data do scale satisfactorily confirming the soundness of our approach. It is clear that, overall, 2D scaling holds much better than for $d=3$, except in a very narrow region at the peak of the scaling function. We believe that this peak is due to three-dimensional fluctuations. This confirms the expectation that intrinsically the 1:2:3 compound is a 2D system in zero field, but in the immediate vicinity of the transition, even though the interlayer coupling is weak, the 3D fluctuation will eventually dominate. For $LuBa_2Cu_3O_{7-y}$ the width of the 3D region can be estimated to be of order unity in the horizontal scale of Fig. 3(b).

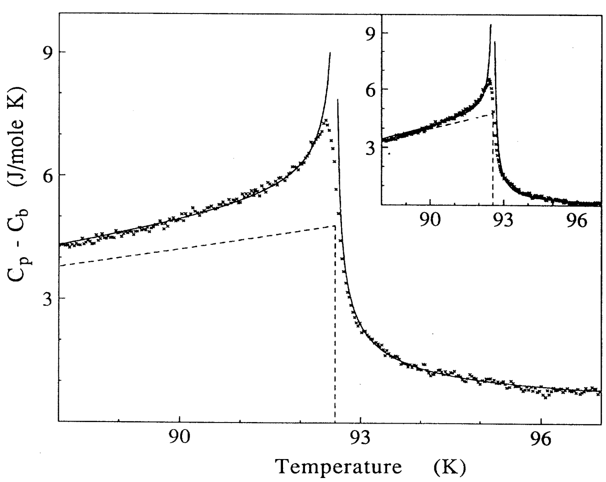


FIG. 2. Zero-field specific heat as a function of temperature over a small temperature window with fitting results. It is clear that the data exhibit a very sharp anomaly. The solid lines are the fitting result to Eq. (1) for $d=3$. The fitting parameters are given in the first row of Table I. (Inset: The fitting result for $d=2$, the parameters being given in the second row of Table I.) The dashed lines are the $C_{\mathrm{MF}}$ term.

A separate question, after this verification of scaling, is the form of the scaling function $g_2$. There are two theoretical predictions. The results of Ref. 24 (based on perturbation theory) and that of the nonperturbative calculation of Ref. 25 are shown in Fig. 3(b) as the dashed and solid lines, respectively. The scaling function of Ref. 25 has the form

$$\Delta C/C_{\mathrm{MF}}=[1-x/(x^2+2)^{1/2}]/2\ , \qquad (3)$$

where $x$ is the argument of $g_2$ in Eq. (2). It is seen that this theoretical curve, without adjustable parameters, fits the experimental results quite well except where the system seems to cross over to 3D behavior. For Ref. 24 there is a factor of order unity relating the scaling variable to the argument of $g_2$.[26] If this factor is taken to be unity, the result shown in Fig. 3(b) is obtained which also

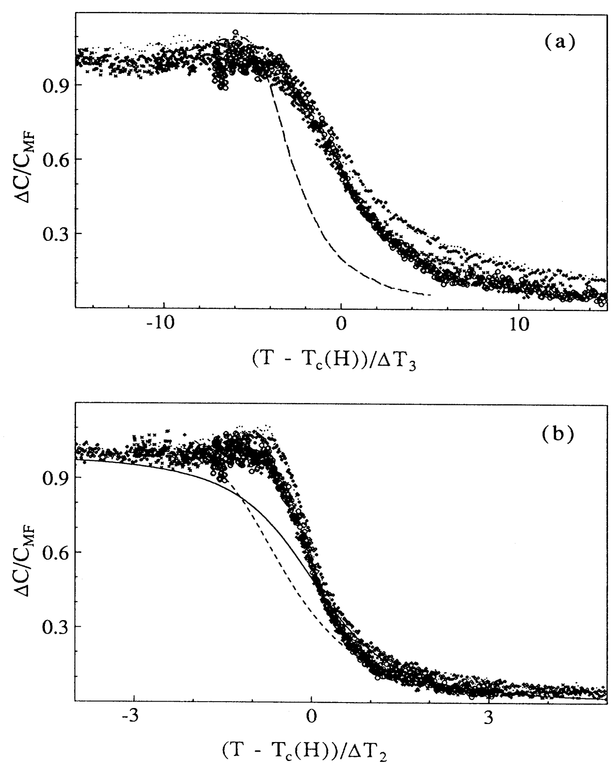


FIG. 3. Scaling plots for $\Delta C/C_{\mathrm{MF}}$ vs $[T-T_c(H)]/\Delta T_d$ of our specific-heat data for $d=3$ (a) and $d=2$ (b), respectively. In (a), the theoretical model by Scalapino, Sears, and Ferrell (Ref. 27) is shown and in (b), the models by Deutscher, Imry, and Gunther (Ref. 24) and Tesanovic *et al.* (Ref. 25) are shown as dashed and solid lines, respectively. The symbols for each field are the same as in Fig. 1.

agrees well with experiment in the 2D region. Other choices, however, lead to less favorable agreement. In Fig. 3(a), we have also included the theoretical curve for a 3D system.[27] Clearly this curve does not accurately describe the system except possibly in the region of the peak. The results shown constitute the first direct evidence of dimensional crossover as function of temperature in the specific-heat measurements of HTSC's.

In conclusion, high-resolution specific-heat data have been acquired from an untwinned $LuBa_2Cu_3O_{7-y}$ single crystal. The finite-field data suggest that the zero-field specific-heat data away from the transition region are better described by a two-dimensional model. So far we do not have any theoretical model, characterizing the crossover behavior from two-dimensional fluctuations to three-dimensional ones, which can well describe our set of specific-heat data as a function of temperature and magnetic field.

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[13]To get an objective comparison among various samples and to minimize uncertainties caused by any extrapolation scheme, we define the size of specific-heat anomaly ($\Delta C_J$) to be the difference between maximum and minimum near the transition temperature. Accordingly, the transition width ($\Delta T_w$) is defined with $\Delta C_J$ being this specific-heat jump.

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[19]The lattice parameters of another $LuBa_2Cu_3O_{7-y}$ single crystal were measured by x-ray diffraction. They are $a=3.821$ Å, $b=3.885$ Å, and $c=11.692$ Å, which are very similar to the values for $YBa_2Cu_3O_{7-y}$.

[20]We have also tried a logarithmic singularity and the interpolation approach of Ref. 9. These models were ruled out for various reasons as will be explained in a paper to follow.

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