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LETTER TO THE EDITOR

Specific heat of an untwinned $YBa_2Cu_3O_{7-x}$ crystal: crossover to critical fluctuations

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Abstract. We present results for the specific heat of an untwinned $YBa_2Cu_3O_{7-x}$ single crystal. The specific heat was measured with a resolution better than 50 mK between 85 K and 110 K. The divergence in the specific heat at T_c is extremely sharp with no indication of rounding. It is not possible to fit the divergence by simply adding Gaussian fluctuations to a BCS-like step, however, we find that the divergence does fit a logarithmic divergence very well over nearly three decades between $10^{-4} < |t| < 0.1$. This can be interpreted either as providing evidence for local pair superconductivity or for a wide crossover region from Gaussian to critical fluctuations.

The exact nature of the specific heat anomaly is of crucial importance in understanding the superconducting transition in high-T_c superconductors. Unfortunately it is an extremely difficult measurement, particularly for small single crystals, which are important to use if we wish to have data on homogeneous samples. The difficulties mean that the published data on the specific heat (C_p) are quite varied, with the specific heat jump (ΔC) ranging from 2 to 8 mJ $g^{-1}K^{-1}$ (Junod 1989). Various authors have reported Gaussian fluctuation contributions to the specific heat in both single crystals and polycrystalline samples (Inderhees *et al* 1988 and 1991, Schnelle *et al* 1990) although Schnelle *et al* (1990) point out that their data fit a logarithmic divergence for $0.01 < |t| < 0.1$; the lower limit on t is probably due to inhomogeneity in the polycrystalline samples used. In this letter we wish to present specific heat data on a homogeneous untwinned single crystal of $YBa_2Cu_3O_{7-x}$, which shows a clear logarithmic divergence to within $t \approx 10^{-4}$ of the transition.

The sample was grown at the University of Leeds using a self-flux technique. The sample was annealed for approximately 14 days at 500 °C in flowing oxygen. Loram *et al* (1991) found that annealing around 500 °C led to the highest T_c with an x of about 0.05 and the sharpest specific heat jumps in the polycrystalline samples. For smaller x they found a double transition and for larger x the jump was very much reduced. The sample originally measured 1.2 mm × 1.2 mm × 50 μm with a large untwinned region in its centre measuring 1 mm × 1 mm and this crystal was used to measure the thermopower in the a- and b-axis directions (Lowe *et al* 1991). A small piece weighing approximately 30 μg was taken from the untwinned region of the crystal for these specific heat measurements. We believe that the long anneal of such a small untwinned crystal has produced a high-quality homogeneous crystal. The heat capacity was measured using a standard

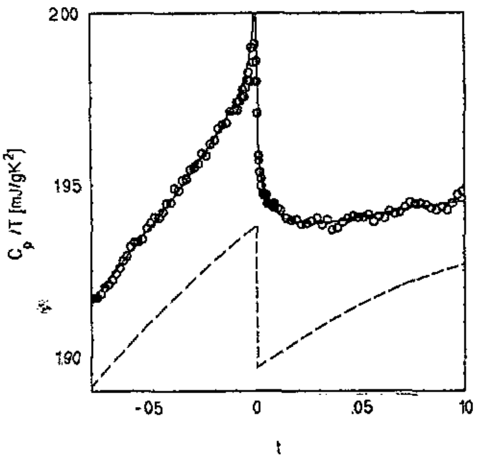

Figure 1. C_p/T data for the $YBa_2Cu_3O_{7-x}$ untwinned crystal. The full curve is the complete fit to the data with a logarithmic divergence. The broken curve is the background with the mean field jump.

AC technique similar to the technique used by the Illinois group (Inderhees *et al* 1991) but using an optical fibre and ≈100 μW LED as the light source.

The specific heat results are shown in figure 1. A very strong positive curvature in the specific heat can be seen close to T_c on both sides of the transition. The temperature oscillations of the crystal are about 15 mK but the data have been collected every 10 mK and averaged over 50 mK close to T_c and over 200 mK well away from T_c. The measured transition is very sharp with an apparent cusp at a T_c of 92.36 K.

The Gaussian fluctuation contribution to the specific heat diverges as a $t^{-1/2}$ law, when $t \gg t_G$, the Ginzburg criterion temprature. For $t < t_G$ the divergence is critical and logarithmic. We have thus tried to fit our data to both a $t^{-1/2}$ and a $\ln(t)$ divergence. The fit to the data was carried out using a quadratic function for the background, along with either a $\ln(t)$ or $t^{-1/2}$ divergence and including a mean field specific heat jump of the form $\Delta C = 1.43\gamma T_c(1 + 1.83t)$ using a non-linear weighted least-squares-fit routine. The backgrounds were then subtracted from the data and the data plotted as a log/log plot for the power law fit in figure 2 and a linear/log plot for the logarithmic fit in figure 3. It is clear from figure 2 that the data fits a $t^{-1/2}$ law over only a very narrow range of t. However, figure 3 shows that the data diverge logarithmically over nearly three decades of t from 0.1 to 10^{-4}, with similar slopes above and below T_c. This is exactly what is predicted for the critical region. In the critical regime the C_p diverges as (Bulaevskii *et al* 1988, Kulic and Stenschke 1988):

$$C_{fl} = -C_0 \ln(t) \qquad t > 0$$

$$C_{fl} = -C_0 \ln(t) + \Delta C \qquad t < 0.$$

Where the ΔC is of the same order, but has a value less than the mean field value, $C_0 = (4k_B/21\pi^2)(1/\xi_0)^3$, and $\xi_0 = (\xi_a\xi_b\xi_c)^{1/3}$. The value of C_0 from the fit is 0.82 mJ g^{-1}K^{-1} and this gives a coherence length ξ_0 of 3.7 Å. This is a very small coherence length; however it is consistent with a wide critical region although even with this value of ξ_0 the Ginzburg temperature t_G is only 5×10^{-3}. The value is smaller than that estimated by Welp *et al* (1989) who found ξ_{ab} to be 16 Å and ξ_c to be 3 Å, so that ξ_0 would be 9 Å. However these estimates are based on estimates of $H_{c2}(0)$ from $dH_{c2}(T)/dT|_{T=T_c}$ measurements and may not be reliable. The value of $\Delta C(T_c)$ obtained from the fits is 3.8 mJ g^{-1}K^{-1}.

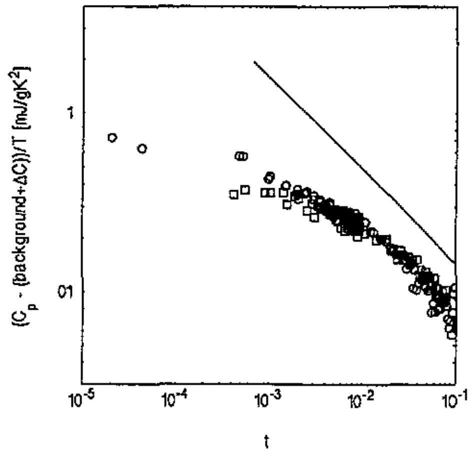

Figure 2. A log/log plot of C_{fl}/T having removed the background from the C_p/T data showing evidence of mean field fluctuations over a narrow range of t. The full curve is a $t^{-1/2}$ slope. □, data below T_c; ○, data above T_c.

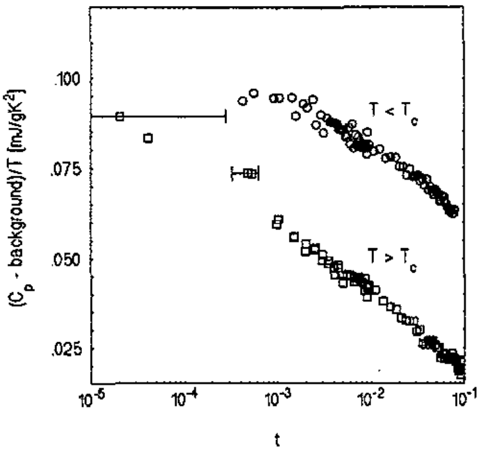

Figure 3. A linear/log plot of C_{fl}/T having removed the background. This shows a very good logarithmic dependence of C_{fl}/T over nearly three decades of t.

We can do a similar analysis on the $t^{-1/2}$ fit. Here we expect the fluctuation contribution to C_p to be of the form $C^{\pm}t^{-1/2}$ where $C^{+} = nk_B/16\pi\xi_0^3$ and the + and − refer to above and below T_c respectively. Here n is the number of components of the Ginzburg–Landau order parameter. We find that the best fit over the limited range $0.01 < t < 0.1$, where a $t^{-1/2}$ fit is appropriate, is obtained for $C^{+} = C^{-}$ which suggests an n of approximately 3. If $n = 2$ we can produce a reasonable fit but the sum of the residuals is 30% smaller for $n = 3$—this may not be significant. The value of C^{+} is 0.23 mJ g^{-1}K^{-1} and this leads to a value of ξ_0 of 8.2 Å. The BCS jump for this fit is 3.9 mJ g^{-1}K^{-1}. All these parameters are similar to other published values (Inderhees *et al* 1991) the significant difference is that the fit is only good to within 0.01 of T_c, while the log fit is good to within 0.0001 of T_c.

These data raise three important issues: the width of the critical regime, the value of the coherence length and the possibility of local pair superconductivity, all of which are closely related. There has been much discussion in the literature concerning the width of the critical regime as defined by the Ginzburg temperature t_G. Estimates vary from 10^{-4} to 0.1 (Fisher *et al* 1991, Sokolov 1991, Bulaevskii *et al* 1988, Kulic and Stenschke 1988). The different estimates arise because of the ambiguity in the definition of t_G and its strong dependence on ξ_0. There has also been some discussion of the possibility of local pair superconductivity in the oxide superconductors (Micnas *et al* 1990, Alexandrov and Ray 1991, Mott 1991). These two questions are interelated since the effective coherence length for local pair superconductors will be a few angstroms and consequently the critical regime will be extremely wide. The problem seems to be that there is not a clear distinction in the properties of local pair and Cooper pair superconductors once the coherence length approaches interatomic dimensions. One feature of the specific heat that is considered strong evidence for a local pair mechanism is the size of the jump in the specific heat $YBa_2Cu_3O_{7-x}$. The size of the jump $\Delta C/k_B$ is related to the number of carriers taking part in the transition. Here, this is the same order of magnitude as the carrier density estimated from Hall effect data and suggests that nearly all the carriers are involved in the phase transition. This would seem reasonable in a local pair model but the BCS theory usually only involves those carriers close to E_F. However in other oxide superconductors the jump may be an order of magnitude smaller than in YBCO, and furthermore, the jump in $YBa_2Cu_3O_{7-x}$ drops rapidly as x is increased while T_c is largely unaffected (Loram *et al* 1990, Mott 1990). The other evidence for a local pair mechanism is a λ-like specific heat anomaly with a wide logarithmic divergence. This is what we have observed in the data presented here, however the logarithmic divergence may simply be evidence for a rather wider critical regime than predicted by the Ginzburg criterion.

It is clear that a critical $\ln(t)$ divergence fits the data for $|t| < 0.1$ but it could be argued that it is difficult to distinguish between $\ln(t)$ and $t^{-1/2}$ for $t > 0.01$. This would suggest that the critical region is around $t < 0.01$. The data show some evidence of mean field $t^{-1/2}$ fluctuations between $t = 0.01$ and 0.1 which supports this possibility. However it must be emphasized that the sum of the residuals for the logarithmic fit are an order of magnitude better than for the $t^{-1/2}$ fit and this implies that the critical region is considerably larger than 0.1. The fact that these values are larger than the estimated t_G of 10^{-4} may not be that surprising (Fisher *et al* 1991). The standard Ginzburg criterion will under-estimate the size of the critical region since at this point the higher-order corrections will be larger than the leading terms in the free energy expansion. However, if the critical region is much larger than 0.1 this—along with the short coherence length of 3.7 Å estimated from the slope of the specific heat—might be considered strong evidence for a local pairing mechanism.

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