Critical and Gaussian fluctuation effects in the specific heat and conductivity of high-\(T_c\), superconductors

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Critical and Gaussian fluctuation effects in the specific heat and conductivity of high-$T_c$ superconductors

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ABSTRACT

We report measurements of specific heat and electrical resistivity for three high-$T_c$ oxides, $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ for $0 \leq \delta \leq 0.18$, $(\text{Ca}_{0.8}\text{Y}_0.2)\text{Sr}_2(\text{Ti}_{0.5}\text{Pb}_{0.5})\text{Cu}_3\text{O}_7$ and $\text{Tl}_{1.8}\text{Ba}_2\text{Ca}_{2.2}\text{Cu}_3\text{O}_{10}$ for which superconducting fluctuations increase rapidly as the interplane coupling is reduced. The coherence lengths $\xi_{ab}$ and $\xi_c$ and Ginzburg temperatures $\tau_G$ are deduced from the data. For $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ we observe a factor of three decrease in $\xi_c$ and an order of magnitude increase in $\tau_G$ between $\delta=0$ and 0·1, resulting in a progression from relatively weak three-dimensional fluctuations for $\delta=0$ to strong two-dimensional (2D) critical fluctuations over a wide temperature range for $\delta>0.1$. Large values of $\tau_G$ and strong 2D critical behaviour are also observed in the other two compounds. We discuss the relevance of these results to the spin-gap behaviour observed above $T_c$ in other properties of oxygen-depleted $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$. Our values of $\xi$ are lower than those deduced from $H_{c2}$ measurements, raising the possibility that additional degrees of freedom contribute to the specific heat.

§1. INTRODUCTION

Understanding the origin of the high transition temperatures $T_c$ in the superconducting cuprates is one of the most fascinating problems in solid-state physics. As discussed for example by Bulaevskii, Ginzburg and Sobyanin (1988), Deutscher (1988) Ginsberg (1988), Lobb (1987) and Salamon (1988), fluctuation effects are much more significant in high-$T_c$ oxides than in conventional superconductors (Skocpol and Tinkham 1975) because of their anisotropic layer structures, lower carrier densities and high values of $T_c$. In this paper we report a combined study of electrical conductivity $\sigma$ and high-resolution specific heat $C$ for three representative high-$T_c$ oxides. There is clear evidence for increasing fluctuation effects as the effective dimensionality, that is the coupling between CuO$_2$ planes, is decreased. These effects are discussed in terms of Ginzburg–Landau (GL) theory using results reviewed by Bulaevskii et al. (1988) with particular emphasis on the determination of coherence lengths and the extent of the critical region. Experimental investigations of fluctuation effects in the specific heat are reviewed by Junod (1989).

Fluctuation effects in high-$T_c$ oxides may turn out to be crucial both with regard to their normal-state properties and for the superconducting state. For example as discussed here they could be connected with the ‘spin gap’ observed above $T_c$ by neutron scattering (Rossat-Mignod et al. 1991), nuclear magnetic resonance (NMR) studies (Berthier et al. 1991) and static susceptibility (Nakazawa and Ishikawa 1989). They may also be relevant to the magnetic-field-induced broadening of the resistive transitions at $T_c$ and other features of the superconducting state such as the
irreversibility line which are not well understood. In principle the possibility of a multicomponent order parameter can be investigated by comparing the fluctuation contributions to $C$ above and below $T_c$.

It is a pleasure to dedicate this paper to Bryan Coles whose insight and enthusiasm have remained an inspiration to those of us (J.W.L. and J.R.C.) fortunate enough to have worked with him.

§ 2. EXPERIMENTAL DETAILS

All the work reported here was made using sintered samples prepared by solid-state reaction. $C$ measurements were made on 2–4 g pellets using a high-resolution differential technique (Loram 1983) and an appropriate reference sample, and resistivity measurements were made on bars cut from the pellets used for the specific-heat measurements. Results for the specific heat (a bulk property) of sintered samples (agglomerates of single crystals) are in no way inferior and are generally more reproducible than those on specially prepared single crystals (Junod 1989), largely on account of the difficulty in controlling the latter's oxygen stoichiometry. To draw fundamental conclusions from the resistivity $\rho(T)$ of sintered samples it is necessary to establish whether $\rho(T) = f \rho_{ab}(T)$ where $f$ is a constant and $\rho_{ab}(T)$ is the a-b-plane resistivity of the corresponding single crystal. There is evidence that this is true for samples of low extrapolated residual resistivity as is the case for the present samples. Experimentally (Babić, Marohnić, Prester and Brnićević 1987, Cooper et al. 1991b) it is found that the resistivity of a sintered sample is strongly dependent on its bulk density (porosity) and the results of the latter paper can be used to estimate the factor $f$ if the bulk density is known.

§ 3. RESULTS AND GENERAL DISCUSSION

Figure 1 shows $C$ data for three oxides near the optimum hole content for maximum $T_c$ of that particular family. The materials are YBa$_2$Cu$_3$O$_{6.95}$ (curve 1), (Ca$_{0.8}$Y$_{0.2}$)Sr$_2$(Tl$_{0.5}$Pb$_{0.5}$)Cu$_2$O$_7$ (curve 2) and Tl$_{1.8}$Ba$_2$Ca$_{2.2}$Cu$_3$O$_{10}$ (curve 3) (hereafter abbreviated to YBCO$_{6.95}$, TIPb and Tl: 2223 respectively). ($C$ is expressed per gram-atom, where 1 g-atom = 173 mol for YBCO$_{6.95}$ and TIPb and 1 g-atom = 173 mol for Tl: 2223 and occupies about 8 cm$^3$ for all three compounds.) Corresponding normalized resistivity plots are shown in fig. 2 with slightly overdoped fully oxygenated YBa$_2$Cu$_3$O$_7$ (YBCO$_7$) (curve 4) included for comparison. The room-temperature resistivities are 0.8, 3.48, 3.47 and 0.6 m$\Omega$ cm and the densities are 86, 50.3, 42 and 86% for YBCO$_{6.95}$, TIPb, Tl: 2223 and YBCO$_7$ respectively. From the results of Cooper et al. (1991b) we estimate $f \approx 4$ for YBCO$_{6.95}$ and YBCO$_7$ and $f = 10–20$ for TIPb and Tl: 2223.

The specific heat data in fig. 1 shows an increase in the fluctuation component above $T_c$ relative to the mean-field step at $T_c$ (asymmetry of the anomaly) from YBCO$_{6.95}$ to TIPb to Tl: 2223. The temperature $T_G$ at which the fluctuation specific heat equals the step marks the boundary between small Gaussian fluctuations and large critical fluctuations. Inspection of fig. 1 shows that the critical region is less than 1 K wide for YBCO$_{6.95}$, 5–10 K wide for TIPb and 10–20 K wide for Tl: 2223. These estimates are confirmed by the more detailed analysis below. A similar trend is observed in the resistance curves in fig. 2 which show a negative curvature characteristic of superconducting fluctuations over a temperature range above $T_c$ of about 20 K for YBCO$_{6.95}$, about 80 K for TIPb and about 150 K for Tl: 2223. These findings indicate a decrease in the effective dimensionality of the superconducting order
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Fig. 1

Specific-heat data for three-$T_c$ oxides, (1), (2) and (3) near the optimum doping for maximum $T_c$.

Fig. 2

Normalized resistivity data for the samples in fig. 1 and including data for YBCO, (4).
parameter from YBCO$_{6.95}$ to TlPb to Tl:2223, implying a reduced coupling between sets of CuO$_2$ planes. The spacing between pairs of CuO$_2$ planes is 7.6 Å (Hazen 1989) and 7.9 Å (Liu et al. 1990) for YBCO$_{6.95}$ and TlPb respectively, while for Tl:2223 it is 8.8 Å between the triple sets of CuO$_2$ planes (Hazen 1989). These minor differences in separation seem insufficient to account for the changes we observe and we believe that the conducting CuO chains in fully oxygenated YBCO play a crucial role in enhancing the interplanar coupling for that system. The data in fig. 1 give clear evidence for a systematic increase in superconducting fluctuations with decreasing interplanar coupling in a model-independent way. In the following we describe a more detailed GL analysis yielding values for coherence lengths and critical temperatures (see also Cooper, Obertelli, Carrington and Loram (1991a) and Loram, Cooper and Mirza (1991b).

§4. FLUCTUATION ANALYSIS

4.1. Sources of uncertainty

When analysing specific heat data, the main problem is the subtraction of the very large phonon contribution. For YBCO$_{7-\delta}$ this has been largely overcome by differential measurements against a fully oxygenated zinc-doped YBCO reference sample, in which superconducting correlations are suppressed with little change in phonon and normal-state electronic specific heat (Loram, Mirza and Freeman 1990). Changes in the phonon term with $\delta$ in YBCO$_{7-\delta}$ can be reliably determined (Loram, Mirza, Cooper and Liang 1992) and the fluctuation term established with confidence over a wide temperature range. Lack of suitable reference samples has so far prevented as complete a cancellation of the phonon term for other superconducting series and investigation of the fluctuation term is restricted to a narrower temperature range about $T_c$.

The fluctuation component of the conductivity $\sigma'$ is determined from $\sigma' = 1/\rho - 1/\rho_n$ where $\rho$ and $\rho_n$ are the total and normal-state resistivities respectively. Following usual practice we assume that $\rho_n$ is linear (or slowly varying) in $T$ and that $\sigma'$ diverges at $T_c$ and is negligible above $2T_c$. Clearly $\sigma'$ becomes very sensitive to the assumed form for $\rho_n(T)$ when $\sigma' \rho_n \ll 1$. A further complication is the possibility of a strongly temperature-dependent Maki–Thompson contribution to $\sigma$ which arises because the scattering probability of a normal carrier is reduced by the presence of superconducting fluctuations. Experimental evidence for this term has been found in studies of zinc-doped YBCO$_{7-\delta}$ (Fung and Cooper 1991, unpublished). The Maki–Thompson term formally diverges at a temperature below $T_c$ dependent on the pair breaking rate, and the Gaussian fluctuation term (see eqn. (2)) dominates at reduced temperatures $\tau < k_B T \tau_{\phi}/\hbar$ where $\tau_{\phi}$ is the pair lifetime.

4.2. Ginzburg–Landau expressions for small Gaussian fluctuations

The GL expressions for the contributions from small Gaussian fluctuations to the specific heat per unit volume $C'$ (Tewordt, Fay and Wolkhausen 1988) and conductivity $\sigma'$ (Lawrence and Doniach 1971) for a layered superconductor above $T_c$ are

$$C' = \frac{n k_B}{2 \sqrt{8\pi}} (\xi_0 \xi d)^{-1} (\tau^2 + \tau \tau^*)^{-1/2}$$  \hspace{1cm} (1)

and

$$\sigma' = \frac{e^2}{32\hbar} d^{-1} (\tau^2 + \tau \tau^*)^{-1/2}$$  \hspace{1cm} (2)
where
\[ \tau^* = \left( \frac{\xi_e}{d} \right)^2. \]

In these expressions \( \xi_a \), \( \xi_b \) and \( \xi_c \) are the \( T=0 \) coherence lengths for the in-plane (a and b) and out-of-plane (c) directions and their product is the coherence volume \( \Omega \), \( \tau \) is the reduced temperature \( (T - T_\theta)/T_c \) where \( T_c \) is the mean-field transition temperature, \( k_B \) is Boltzmann's constant and \( 2d \) is the average spacing between independently fluctuating \( \text{CuO}_2 \) planes. \( n \) is the number of independent components of the order parameter; \( n = 2 \) (amplitude and phase) for conventional and most proposed unconventional superconductors and we assume this to be the case unless otherwise stated.

Equations (1) and (2) reduce to the standard expressions for three-dimensional (3D) and two-dimensional (2D) fluctuations in the appropriate limits:

\[ 3D, \quad \tau \ll \tau^* \quad C' = \frac{k_B}{8\pi} \Omega^{-1} \tau^{-1/2}, \quad (3) \]
\[ \sigma' = \frac{e^2}{32\hbar} \xi_e^{-1} \tau^{-1/2} \quad (4) \]

and

\[ 2D, \quad \tau \gg \tau^* \quad C' = \frac{k_B}{8\pi} \left( \xi_{ab}d \right)^{-1} \tau^{-1}, \quad (5) \]
\[ \sigma' = \frac{e^2}{32\hbar} d^{1/2} \tau^{-1} \quad (6) \]

and according to eqns. (1) and (2) there is a cross-over from 3D \((C', \sigma' \propto \tau^{-1/2})\) to 2D behaviour \((C', \sigma' \propto \tau^{-1})\) above the reduced temperature \( \tau^* = (\xi_e/d)^2 \).

Formulae for \( \sigma' \) have also been derived for the physically realistic case where there are pairs of planes (as for YBCO) and two interplane coupling constants (Maki and Thompson 1989). At low values of \( \tau \), each pair of planes will fluctuate as one unit and \( 2d \) will correspond to the c-axis lattice parameter, but at high \( T \) the planes decouple and the effective value of \( 2d \) decreases by a factor of two. We assume the former case throughout and take \( 2d = 11.7, 12.1 \) and 18 Å for YBCO\( _{6.95} \), TlPb and Tl:2223 respectively.

4.3. Results from a GL analysis

The parameters derived from an analysis of the specific-heat data in terms of eqn. (1) are summarized in the table. However, several significant trends may be deduced by comparing the data with the simplified expressions eqns. (3) and (5). Differences in effective dimensionality for YBCO\( _{6.95} \), TlPb and Tl:2223 are immediately apparent from the plots of \( 1/\Delta C \) against \( T \) shown in fig. 3. \( \Delta C \) is the excess specific heat after subtraction of a normal state background, and \( \Delta C = C' \) for \( T > T_c \). For YBCO\( _{6.95} \) an initial \((T-T_\theta)^{1/2}\) dependence confirms 3D fluctuations close to \( T_c \) with a gradual cross-over to 2D behaviour at higher temperatures (see also the \( 1/\Delta C^2 \) plot in fig. 5(a)). By contrast the Tl:2223 sample exhibits linear behaviour until very close to \( T_c \) consistent with a cross-over from 2D to a critical fluctuation region. (Note that the transition for this sample is too sharp (about 1-2 K) to attribute the lack of curvature to sample inhomogeneity. A spread of \( T_c \) of this magnitude is sufficient, however, to suppress a weak logarithmic divergence.)
Values derived from a GL analysis of the fluctuation specific heat; the coherence volume $\Omega$ in $\text{Å}^3$, the coherence lengths $\xi_{ab}$ and $\xi_e$ in $\text{Å}$, the 3D-to-2D cross-over temperature $t^*$ and the parameters $\tau_{G,2D}$ and $\tau_G$ related to the critical region (§4.4). Note that the values given for $\xi_{ab}$ and $\xi_e$ of the more 2D compounds should be treated with caution because they obtained in a temperature range where the condition $t \gg \tau_G$ is clearly not satisfied. Values of $\xi_e$ and $t^*$ from the fluctuation conductivity are given in the last two columns (from Cooper et al. (1991a)).

<table>
<thead>
<tr>
<th>Compound</th>
<th>Abbreviation</th>
<th>$\Omega$</th>
<th>$\xi_{ab}^2$</th>
<th>$\xi_e$</th>
<th>$t^*$</th>
<th>$\tau_{G,2D}$</th>
<th>$\tau_G$</th>
<th>$\xi_e$</th>
<th>$t^*$</th>
</tr>
</thead>
<tbody>
<tr>
<td>YBa$_2$Cu$_3$O$_7$</td>
<td>YBCO$_7$</td>
<td>400</td>
<td>125</td>
<td>3.2</td>
<td>0.30</td>
<td>0.022</td>
<td>0.0016</td>
<td>2.8</td>
<td>0.23</td>
</tr>
<tr>
<td>YBa$_2$Cu$<em>3$O$</em>{7-\delta}$, $\delta=0.025$</td>
<td>YBCO$_7-\delta$</td>
<td>309</td>
<td>119</td>
<td>2.6</td>
<td>0.20</td>
<td>0.023</td>
<td>0.0025</td>
<td>2.3</td>
<td>0.16</td>
</tr>
<tr>
<td>$\delta=0.05$</td>
<td></td>
<td>250</td>
<td>119</td>
<td>2.1</td>
<td>0.13</td>
<td>0.023</td>
<td>0.0038</td>
<td>1.8</td>
<td>0.1</td>
</tr>
<tr>
<td>$\delta=0.1$</td>
<td></td>
<td>143</td>
<td>119</td>
<td>1.2</td>
<td>0.042</td>
<td>0.037</td>
<td>0.021</td>
<td>1.4</td>
<td>0.06</td>
</tr>
<tr>
<td>Ca$<em>{0.8}$Y$</em>{0.2}$Sr$<em>2$Tl$</em>{0.5}$Ph$_{0.5}$Cu$_2$O$_7$</td>
<td>TIPb</td>
<td>84</td>
<td>70</td>
<td>1.2</td>
<td>0.045</td>
<td>0.12</td>
<td>0.1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Tl$_{1.8}$Ba$<em>2$Ca$</em>{2.2}$Cu$<em>5$O$</em>{10}$</td>
<td>Tl: 2223</td>
<td>40</td>
<td>&lt;0.9</td>
<td>&lt;0.01</td>
<td>0.16</td>
<td>0.16</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
The excess specific heat $\Delta C$, after subtraction of the phonon and normal carrier background, is plotted as $1/\Delta C$ against $T$ for the samples in fig. 1. 3D and 2D fluctuations yield $1/\Delta C \propto (T - T_c)^{1/2}$ and $1/\Delta C \propto T - T_c$ respectively.

Specific-heat data for YBCO$_{7-\delta}$ with $0 \leq \delta \leq 0.18$. A constant normal-state background term of about 1.6 mJ/(g-atom)$^{-1}$ K$^{-2}$ has been subtracted.
Figure 4 shows the specific heat anomalies at $T_c$ for a YBCO$_{7-\delta}$ sample with $\delta$ varied by annealing in an oxygen atmosphere at appropriate temperatures (the $\delta$ dependence of the resistivity, $\sigma'$, thermopower and Hall coefficient data for this sample have been described by Cooper et al. (1991a) and the splitting of the specific-heat transition for very low $\delta$ has been discussed by Loram et al. (1991b)). Fluctuations above $T_c$ grow rapidly with increasing $\delta$ and for $\delta=0.18$ the anomaly closely resembles

Fig. 5

(a) The specific-heat data shown in fig. 4 for YBCO$_{7-\delta}$ with $\delta=0-0.1$, plotted as $1/\Delta C^2$ against $T$. For 3D fluctuations, $1/\Delta C^2 \propto \Omega^2(T - T_c)$, where $\Omega$ is the coherence volume. (b) The fluctuation conductivity for YBCO$_{7-\delta}$ with $\delta=0-0.18$ plotted as $1/\sigma'^2$ against $T$. For 3D fluctuations, $1/\sigma'^2 \propto \xi_c(T - T_c)$ where $\xi_c$ is the c-axis coherence length (Cooper et al. 1991a).
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those for TIPb and Tl:2223 (fig. 1). Plots of $1/\Delta C^2$ against T and $1/\sigma^2$ against T are shown if figs. 5(a) and (b) respectively for $0 \leq \delta \leq 0.1$. Above a temperature region close to $T_c$ to be discussed in the next section, both properties show an initial linear temperature dependence, indicating 3D fluctuations. The decrease in slopes with increasing $\delta$ correspond to a decrease in coherence volume $\Omega$ from the specific-heat data in fig. 5(a) (eqn. (3)) and to a decrease in $\xi_e$ from the conductivity data in fig. 5(b) (eqn. (4)). A detailed analysis of the data in terms of eqns. (1) and (2) gives good agreement between the estimates of $\tau^*$ and $\xi_e$ from both sets of data (table) and confirms that the decrease in $\Omega$ with increasing $\delta$ results from a reduction in $\xi_e$ whilst $\xi_{ab}^2$ remains essentially constant (Cooper et al. 1991a).

4.4. Ginzburg temperatures

Equations (1)–(6) describe small amplitude (Gaussian) fluctuations. As $T_c$ is approached, the fluctuations grow in amplitude and the GL approximation breaks down in the critical region characterized by the Ginzburg temperature $\tau_G$. Values of $\tau_G$ appropriate to 2D and 3D systems are given by the reduced temperatures at which the fluctuation contributions $C'$ from eqns. (5) and (3) equal the mean-field step $\Delta C^{MF}$ at $T_c$. This gives

$$
\tau_{G,2D} = \left( \frac{8\pi \xi_{ab}^2 d \Delta C^{MF}}{k_B} \right)^{-1},
$$

$$
\tau_{G,3D} = \left( \frac{8\pi \Delta C^{MF}}{k_B} \right)^{-2} \frac{\tau_{G,2D}^2}{\tau^*}.
$$

(Bulaevskii et al. 1988) use for the 3D case $C'(\tau_G) = \Delta C^{MF}/2^{1/2}$, yielding values of $\tau_{G,3D}$ a factor two higher than eqn. (8).) For a layered superconductor, $\tau_{G,2D}$ is the critical temperature that the system would have in the absence of interplane coupling, $1/\tau_{G,2D}$ being of the order of 50–100 times the condensation energy $U$ (expressed in units of $k_B T_c$) in a volume $\xi_{ab}^2 d$. The relative magnitudes of $\tau_{G,2D}$ and $\tau^*$ are crucial for assessing the importance of fluctuations. If $\tau_{G,2D} < \tau^*$, then the sample passes through a region of weak 3D fluctuations before entering the critical region below $\tau \approx \tau_{G,3D}$. If $\tau_{G,2D} > \tau^*$, the system progresses directly from 2D Gaussian to critical below $\tau \approx \tau_{G,2D}$. Note that the large numerical factor in the expressions for $\tau_G$ yields very low values for $\tau_G$ even when $U/k_B T_c \approx 1$ and the temperature range over which critical behaviour is significant may be much larger than $\tau_G$. Thus the $\lambda$ anomaly in the specific heat of superfluid $^4$He shows a logarithmic singularity to $\tau \approx 0.05$ (i.e. $T - T_\lambda \approx 0.1$ K) even though $\tau_G \approx 10^{-3}$ (Bulaevskii et al. 1988).

The mean-field step $\Delta C^{MF}$ is determined from the asymmetry of the specific-heat anomaly. When it is clear that the 3D or 2D limits apply, the quantities $\tau_{G,3D}$ or $\tau_{G,2D}$ may be estimated directly from appropriate plots of fluctuation specific heat using

$$
C'_{3D} = \Delta C^{MF} \left( \frac{\tau_{G,3D}}{\tau} \right)^{1/2}
$$

or

$$
C'_{2D} = \Delta C^{MF} \frac{\tau_{G,2D}}{\tau}.
$$
In general they may be obtained from eqns. (7) and (8) with coherence lengths derived from fits of $C'$ to eqn. (1). Values for $\tau_{G,2D}$ so obtained are given in the table. The YBCO samples with low $\delta$ all have $\tau_{G,2D} < \tau^*$ and thus exhibit 3D behaviour close to $T_c$. At the other extreme the TL:2223 sample has $\tau_{G,2D} > \tau^*$ and shows only 2D Gaussian and critical behaviour as noted above. We can estimate $\tau_G$ for intermediate situations using eqn. (1) with $C(\tau_G) = \Delta C_{MF}$. This gives

$$\tau_G = \left[ \tau_{G,2D} + \left( \frac{\tau^*}{2} \right)^2 \right]^{1/2} \frac{\tau^*}{2},$$

(9)

which reduces to $\tau_{G,2D}$ and $\tau_{G,3D}$ in the appropriate limits.

Using eqn. (9) we have determined the values of $\tau_G$ given in the table and these help to explain the variety of behaviours observed in figs. 1–5. The most remarkable feature of the $\tau_G$ values for YBCO$_{7-4}$ is the order-of-magnitude increase in $\tau_G$ between $\delta = 0$ and 01 which may be traced to the factor-of-three decrease in $\xi_e$. This results in a change from a regime of small-amplitude 3D fluctuations for $\delta = 0$ to superconductivity dominated over a wide temperature range by large-amplitude 2D critical fluctuations for $\delta \geq 0.1$. Similar strong 2D critical fluctuations predominate in TlPb and Tl:2223 and from specific-heat evidence may well do so in all other known oxide superconductors. The unique 3D behaviour in fully oxygenated YBCO is, we believe, due to the development of superconducting order on the CuO (chain) planes which increases the coupling between successive pairs of CuO$_2$ planes and thus increases $\xi_e$. These results illustrate an important consequence of eqn. (9) which shows that $\tau_G$ is extremely sensitive to $\tau^*$, decreasing rapidly in the 3D limit when $\tau^* \geq \tau_{G,2D}$. Whereas $\tau_{G,2D}$ is a measure of the amplitude of the order parameter on the CuO$_2$ planes and may show little variation between oxide superconductors, $\tau^* (= \xi_e/d^2)$ depends on the coupling between the planes and shows considerable variation. From eqn. (9) the cross-over from strong 2D critical behaviour to weak 3D behaviour can be achieved with a fairly modest increase in $\xi_e$.

An interesting manifestation of critical behaviour may be seen in fig. 5. The linear region of plots of $1/\Delta C^2$ against $T$ extrapolate to zero at temperatures lower than the measured value of $T_c$ (determined from the temperature of maximum negative slope of the specific heat anomaly), whereas from eqn. 1 they should extrapolate to zero at the mean-field transition temperature. Similar behaviour is observed in plots of $1/\sigma'^2$ against $T$ for these samples (fig. 5(b)) (Cooper et al. 1991a), single-crystal YBCO$_7$ (Friedmann, Rice, Giapintzakis and Ginsberg 1989) and sintered (Ca$_{1-x}$Y$_x$)Sr$_2$(Ti$_{0.5}$Pb$_{0.5}$)Cu$_2$O$_7$ (Loram et al. 1991a) and show that $C'$ and $\sigma'$ diverge more slowly than $\tau^{-1/2}$ (smaller negative power of $\tau$) close to $T_c$. We suggest that these 'negative' intercepts (which are comparable in magnitude with $\tau_G T_c$ from the table) are caused by fluctuation-mode-coupling effects which set in as the Ginzburg temperature $T_G$ is approached, reducing the amplitude of the fluctuations of the order parameter relative to the mean-field value. A perturbation calculation by one of us (J.M.W.) for the 3D O(n) GL model shows that, for $\tau \gg \tau_G$,

$$C' = C'_{3D} \left[ 1 - 3(n+2) \left( \frac{\tau_G}{\tau} \right)^{1/2} + \ldots \right],$$

(10)

where $C'_{3D}$ is given by eqn. (3) and again a more general $n$-component order parameter has been taken into account. Equation (10) confirms that $C'$ is reduced when critical effects are included and qualitatively describes the extrapolation of $1/\Delta C^2$ to an
intercept below $T_n$ but higher-order terms in the expansion are required for quantitative comparison when $\tau < 10^3 \tau_G$. The expression

$$C' = \alpha \ln \left[ 1 + \beta \left( \frac{\tau_G}{\tau} \right)^{1/2} \right]$$

(11)

with $\beta = 6(n + 2)$ and $\alpha = n \Delta C_{MF}^{\text{MF}}/2 \beta$ provides a convenient interpolation between eqn. (10) for $\tau \gg \tau_G$ and the observed logarithmic divergence near $T_c$ (Reagan, Lowe and Howson 1991) and accounts well for the curves in fig. 5(a). When suitably modified to yield eqn. (1) for $\tau \gg \tau_G$, eqn. (11) fits our specific-heat data for both the 3D and the 2D systems described here (Loram et al. 1992).

The depression of $T_c$ below the mean-field value $T_{c0}$ due to 3D critical fluctuations has also been estimated using standard scaling techniques (Ma 1976). The reduced shift in $T_c$ is given by

$$\delta T_c = \frac{T_{c0} - T_c}{T_c} = (n + 2) \frac{A \xi_0}{\pi} \tau_G^{1/2}.$$

(12)

$\xi_0$ is the coherence length and $A \approx 1/\xi_0$ is a momentum space cut-off. With the values of $\tau_G$ in the table, eqn. (12) gives $\delta T_c \approx 0.06$ for YBCO$_{7-\delta}$ with $\delta \leq 0.05$. This agrees with the experimental estimate $\delta T_c = 6$ K by Inderhees, Salamon, Rice and Ginsberg (1991), but the above value of $A$ is a lower limit. We would expect the depression of $T_c$ to be substantially larger for the more 2D samples with larger values of $\tau_G$.

§ 5. DISCUSSION

It is interesting to note that, within a single-band picture, the large 2D superconducting fluctuations associated with the high values of $\tau_G$ that we observe in oxygen-deficient YBCO$_{7-\delta}$ may account for the strongly temperature-dependent spin susceptibility (Nakazawa and Ishikawa 1989) and spin-gap effects observed in neutron scattering (Rossat-Mignod et al. 1991) and NMR studies (Berthier et al. 1991) to temperatures well above $T_c$. This follows from the fact that, as $T$ is reduced towards $T_c$, the loss in entropy associated with the fluctuations becomes comparable with the entropy of the carriers in the normal state, generating an apparent ‘pseudo-gap’ in the normal-state excitation spectrum. Estimates from the fluctuation specific heat described here and indeed direct determinations of the entropy for oxygen-depleted YBCO$_{7-\delta}$ (Loram et al. 1992) show that for $\delta \geq 0.18$ the entropy at $T_c$ is reduced by fluctuations by 20–50% which is comparable with the fall in the static susceptibility between 400 K and $T_c$ (Nakazawa and Ishikawa 1989). For $\delta \approx 0$, spin-gap effects above $T_c$ disappear (Berthier et al. 1991) and $\chi$ is temperature independent down to $T_c$, consistent with our observation that, for $\delta < 0.1$, $\tau_G$ and the entropy of the fluctuations above $T_c$ decrease rapidly.

A significant problem remains in that the value of $\tau_G$ for YBCO$_7$ from our specific-heat measurements (about $2 \times 10^{-3}$) is substantially higher than that of Bulaevskii et al. (1988) who estimate $\tau_G \approx 2 \times 10^{-5}$ – $2 \times 10^{-4}$. This can be traced to their determination of $\tau_G$ from coherence lengths deduced from $H_{c2}$ measurements which yield $\Omega \approx 4500$ or 1700 Å$^3$, an order of magnitude larger than our specific-heat values (table). Indeed our values of $\xi_{ab}$ and $\xi_c$ correspond to $H_{c2}^0(0) \approx 320$ T and $H_{c2}^0(0) \approx 1000$ T, substantially higher than values estimated from $dT_c/dH$ close to $T_c$. If our specific-heat estimates are correct we must conclude that critical effects increase $(dT_c/dH)_{T_G}$, leading to underestimates of critical fields and overestimates of coherence lengths. If, however, we accept
the higher values of $\xi_{ab}$ and $\xi_{c}$ from $H_{c2}$ measurements we must conclude that there are additional degrees of freedom enhancing the specific heat fluctuations and leading us to underestimate $\xi_{ab}$ and $\xi_{c}$ and overestimate $\tau_{G}$. This could be due to a separation of spin and charge degrees of freedom, to an additional divergent phonon contribution (Obhi and Salje 1990) or to unconventional pairing with $n \geq 2$.

§ 6. SUMMARY AND CONCLUSIONS

In summary we have reported new specific heat and conductivity data for several high-$T_c$ oxides and have made a detailed fluctuation analysis of $C$ data for slightly oxygen-deficient YBCO$_{7-\delta}$. For fully oxygenated YBCO$_7$, the Ginzburg temperature $\tau_{G}$ is relatively small (similar to that for the $\lambda$ transition in $^4$He) and weak 3D fluctuations are observed. Tl$_2$2223 and TlPb and deoxygenated YBCO$_{7-\delta}$ have large $\tau_{G}$ and exhibit strong 2D critical fluctuations. Since the specific-heat anomalies in other oxide superconductors are similar in size and character to those of the latter compounds, we believe that strong 2D critical behaviour may be the norm and that the 3D behaviour of fully oxygenated YBCO may be unique, accounting for its superior technical properties in a magnetic field. We suggest that the trends that we observe in $\tau_{G}$ may account for the so-called spin-gap effects observed in other properties.

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