Specific Heat of YBa$_2$Cu$_3$O$_{7-\delta}$ Single Crystals: Implications for the Vortex Structure

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Abstract

The anisotropy of the magnetic field dependence of the specific heat of YBa$_2$Cu$_3$O$_{7-\delta}$ can be used to identify different low-energy excitations, which include phonons, spin-$\frac{1}{2}$ particles, and electronic contributions. With a magnetic field $H$ applied perpendicular to the copper oxide planes, we find that the specific heat includes a linear-$T$ term proportional to $\sqrt{H}$. The nonlinear field dependence of the density of states at the Fermi level suggests that there are quasiparticle excitations throughout the entire vortex, not just in the vortex core. The $\sqrt{HT}$ term agrees quantitatively with G. Volovik’s prediction for a superconductor with lines of nodes in the gap. A similar, but much smaller, effect is predicted for fields parallel to the planes, and sensitive measurements of the in-plane anisotropic magnetic field dependence of the specific heat could be used to map out the nodes.
1 Introduction

Among recent experiments supporting the possibility of lines of nodes in the gap function of some of the high-\(T_c\) materials [1], measurements on single-crystal YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\) (YBCO) show a linear temperature dependence of the penetration depth [2], as predicted for superconductors with lines of nodes in the clean limit [3]. The density of states \(N(E)\) in such superconductors rises linearly with energy at the Fermi level in zero field, which should result in a quadratic term \(\alpha T^2\) in the specific heat [4]. A material with a finite \(N(E_F)\), such as a normal metal or a disordered superconductor with lines of nodes [5], has a linear term \(\gamma T\) in the zero-field specific heat, with \(\gamma \propto N(E_F)\).

G. Volovik predicted that for a superconducting state with lines of nodes, the dominant contribution to the magnetic field dependence of the density of states at the Fermi level is 
\[
N(E_F, H) = \kappa N_n \sqrt{H/H_{c2}},
\]
where \(N_n\) is the normal-state DOS and \(\kappa\) is a factor of order 1 [6]. The factor \(\kappa\) is defined by the vortex lattice structure, by the slope of the gap near the gap node, and by possible structure in \(N_n\). The quasiparticles which contribute to \(N(E_F, H)\) are outside the vortex cores, and are close to the nodes in momentum space. By contrast, it is usually thought that in a clean s-wave superconductor the quasiparticles are confined to the vortex cores.

In previous papers [8, 9] we reported measurements of the magnetic field dependence of the specific heat of twinned and untwinned single crystals of YBa\(_2\)Cu\(_3\)O\(_{6.95}\) which were prepared identically to those showing linear temperature dependence of the penetration depth [2, 10]. These crystals have a linear term \(\gamma(0)T\) in the zero-field specific heat, a quadratic term \(\alpha T^2\) which is only a few percent of the total zero-field specific heat, and an additional field-dependent linear-T term \(\gamma_\perp(H)T\) which obeys \(\gamma_\perp(H)T = A\sqrt{HT}\). Two of these terms, \(\alpha T^2\) and \(A\sqrt{HT}\), agree quantitatively with the predictions of lines of nodes.

In this paper we summarize the previous analysis, including alternative fits, and present results on the sample dependence of the zero-field linear term, \(\gamma(0)T\). We also discuss the physical origin of the \(\sqrt{H}\) dependence of the density of states and the prospects for observing this effect in a magnetic field applied parallel to the plane. In principle, measurements of the in-plane anisotropic magnetic field dependence of the specific heat could be used to map out the nodes.
2 Technique

The specific heat was measured using a relaxation method described in detail elsewhere [1]. The sample was mounted on a sapphire substrate with a weak thermal link (thermal conductance $\kappa_w$) to a constant-temperature copper block. At each temperature and field, $\kappa_w$ was measured by applying power $P = \kappa_w(T_{\text{sample}} - T_{\text{block}})$. A smaller temperature difference was then used to make a relaxation measurement of the thermal time constant $\tau = C\kappa_w$, where $C$ is the total heat capacity. The precision of the measurement in the temperature range 2–10 K is $\sim 0.5\%$. The addenda heat capacity can be as high as half of the total heat capacity, limiting the absolute accuracy in some measurements to $\sim 10\%$. The addenda specific heat is mostly due to phonons, so that most of the systematic uncertainty effects only the Debye $\beta T^3$ term. Measurements of an empty substrate showed that the addenda heat capacity does not depend on the applied magnetic field within the precision of the data. The accuracy of the field dependence of the specific heat is better than 1%.

3 Crystals

All of the samples are single crystals of YBa$_2$Cu$_3$O$_{7-x \delta}$ grown by the same flux-growth technique [10] as crystals which show linear temperature dependence of the penetration depth [2]. Results have been obtained on two nominally pure, twinned single crystals (samples T1 and T2). A 0.8 mg piece was cleaved from one of the twinned crystals, sample T1, and detwinned to create sample U1. About 20% of the surface of sample U1, near two of the corners, had a high density of visible twins remaining after the detwinning process. Sample T2 was mostly detwinned to create sample U2, which had visible twin boundaries remaining. Both U1 and U2 were reoxygenated subsequent to detwinning.

4 Results

Figure 1 shows the specific heat of sample T1 in an 8 Tesla field applied either parallel or perpendicular to the copper oxide planes. The total specific heat includes the Debye phonon contribution, $\beta T^3$, which has a similar value
in all three data sets; the zero-field linear term $\gamma(0)T$, which appears as a finite intercept on the $c/T$-axis; a magnetic contribution from spin-$\frac{1}{2}$ particles, $nc_{\text{Schottky}}(T, H)$, which appears as a low-temperature bump peaking around 3-4 Kelvin in both parallel and perpendicular fields; and a substantially increased linear term in perpendicular field, $\gamma_\perp(H)T$.

### 4.1 Perpendicular Field—Global Fit

To model the perpendicular field dependence, identical analyses are performed on data from samples T1 ($H = 0, 0.5, 2, 4, 6,$ and 8 Tesla) and U1 ($H = 0, 0.5, 2, 4, 6, 8,$ and 10 Tesla). A global fit is used to constrain the concentration of spin-$\frac{1}{2}$ particles, $n$, and the phonon specific heat, $\beta T^3$, to be field-independent. Allowing for the possibility of a small quadratic term, $\alpha T^2$, as predicted for lines of nodes, the zero-field specific heat is described by:

$$c(T, 0) = \gamma(0)T + \alpha T^2 + \beta T^3.$$  

(1)

The specific heat for $H \geq 0.5$ T is described by:

$$c(T, H) = (\gamma(0) + \gamma_\perp(H))T + \beta T^3 + n c_{\text{Schottky}}(g\mu_B H/k_B T),$$  

(2)

where

$$c_{\text{Schottky}}(x) = x^2 \frac{e^x}{(1 + e^x)^2}.$$  

(3)

All data sets are fit simultaneously to Equations 1, 2, and 3 with a Landé g-factor of $g = 2$. Thus, $\beta$ and $n$ are constrained by multiple data sets, $\gamma(0)$ and $\alpha$ are constrained by the zero-field data, and $\gamma_\perp(H)T$ is allowed a different value at each field. The statistical confidence levels are determined by a bootstrapping technique, in which alternative data sets are randomly chosen from the original data set and the fitting procedure is repeated. This procedure for determining the statistical error bars avoids making any assumption about the statistical error distribution of the data set.

The parameters determined from this fit are shown in Table 1, with 90% statistical confidence levels. The coefficient of the Schottky anomaly, $n$, indicates 0.1% spin-$\frac{1}{2}$ particles per Copper. The coefficient of the Debye term, $\beta = 0.39$ mJ/molK$^4$, corresponds to a Debye temperature of 400 K. The difference in the Debye term between the two samples is within the systematic error of the Debye term of the addenda. The rms deviation of the data from the fit is 0.8% for T1 and 2.7% for U1. The higher scatter in
the data for the untwinned crystal U1 results from the smaller sample size. The data and fits for both samples are shown in Figure 2, with the phonon specific heat ($\beta T^3$) subtracted.

Table 1: Global Fit Parameters (mJ, mol, K, and T)

|       | Sample T1   | Sample U1   |
|-------|-------------|-------------|
| $\gamma(0)$ | $3.0 \pm 0.1$ | $2.1 \pm 0.1 - 0.2$ |
| $n$    | $24 \pm 1$  | $23 \pm 1$  |
| $\beta$ | $0.392 \pm 0.001$ | $0.380 \pm 0.004$ |
| $\alpha$ | $0.11 \pm 0.02$ | $0.10 \pm 0.06$ |
| $\gamma(\perp H)$ | $0.91\sqrt{H}$ | $0.88\sqrt{H}$ |

In the measurements on single crystals of YBCO reported here, the phonon specific heat obeys the Debye $\beta T^3$ law up to 8 K. In the zero-field data set on sample T1, for example, fits from 2 to 4 K and from 4 to 7 K give the same value of $\beta$ within statistical error, indicating that higher powers of temperature are not necessary to describe the data. Above 8 K, the deviations from $\beta T^3$ are better described by a gapped excitation such as an optical phonon mode than by a sum of $T^5$, $T^7$, and $T^9$ terms. In contrast, the data on many polycrystalline samples require a large $T^5$ term to describe the phonon specific heat even below 5 K [13]. In order to avoid the extra parameters necessary to describe the phonon specific heat above 8 K, all of the fits reported in this paper are restricted to $T \leq 7$ K.

The field-dependent linear term, $\gamma(\perp H)T$, has a nonlinear dependence on field (Figure 3). This nonlinear field dependence is well-described by $\gamma(\perp H) = A\sqrt{H}$, with $A = 0.9$ mJ/molK$^2 T^{1/2}$ for both samples T1 and U1.

4.2 Perpendicular Field—Alternative Fits

The qualitative nonlinearity of $\gamma(\perp H)$ is robust to the assumptions used to describe the total specific heat, and is well described by $\gamma(\perp H) = A\sqrt{H}$. To check the assumptions of the global fit and the interdependence of the $\alpha T^2$ and $A\sqrt{HT}$ terms, each data set of sample T1 is fit independently to Equation 2. There are thus three parameters each for five data sets ($H = 0.5$–8 T) and two parameters for the zero-field data set (note that $c_{\text{Schottky}}(T, 0) =$
0), totaling seventeen parameters for six data sets. No zero-field quadratic term was allowed in this fit. A small quadratic term, if present, could easily be absorbed into a slightly larger cubic term, and this fit results in a $\beta(H = 0)$ which is $\sim 5\%$ higher than $\beta(H \neq 0)$. The coefficients of the linear term are shown in Figure 4a with error bars at the statistical 90\% confidence level. Fitting the coefficients of the linear term to $\gamma_{\perp}(H) = A\sqrt{HT}$ gives $A = 0.89$ mJ/molK$^2$T$^{1/2}$.

In another check which avoids the zero-field data entirely, the five data sets with $H \geq 0.5$ T are globally fit to Equation 2, again keeping $\beta$ and $n$ independent of field. There are thus two global parameters, plus values of $(\gamma(0) + \gamma_{\perp}(H))$ in five fields (Figure 4b). The resulting linear-T term is better described by a $\sqrt{H}$ dependence than by an $H$ dependence, and from the fit shown in Figure 4b, $A = 0.87$ mJ/molK$^2$T$^{1/2}$. This fit also returns an extrapolated value of $\gamma(0) = 3.1$ mJ/molK$^2$, which is in agreement with the value determined by independently fitting just the zero-field data.

4.3 Parallel Field

The data on sample T1 in an 8 Tesla parallel field show a linear-T term, $\gamma_{\parallel}(H)T$, which is increased by about 0.5 mJ/molK$^2$ from $\gamma(0)$, as determined by fitting the data in Figure 3 to Equation 2. Because of uncertainties in the Schottky anomaly and the sample alignment, the results are also consistent with no increase in the parallel-field linear term, $\gamma_{\parallel}(H)T$.

4.4 Zero-field Linear Term

The coefficient of the zero-field linear term, $\gamma(0)T$, in the specific heat of each of the four samples is shown in Table 2. The zero-field linear term for the two twinned samples, samples T1 and T2, is $\gamma(0) = 3$ mJ/molK$^2$, while in both untwinned samples, U1 and U2, the linear term is reduced to $\gamma(0) = 2$ mJ/molK$^2$. In contrast, most YBCO samples have $\gamma(0) \geq 4$ mJ/molK$^2$ [13]. The coefficient of the linear term in the normal state of optimally-doped YBCO, $\gamma_{n}$, has been shown by other measurements to be about 20 mJ/molK$^2$ [13, 14]. The residual density of states $N(E_F)$ in the superconducting state is therefore about 15\% of the normal-state density of states for the twinned samples and about 10\% for the untwinned samples.
Table 2: Zero-Field Linear Term, $\gamma(0)$ (mJ/molK$^2$)

| Sample | $7 - \delta$ | Untwinned | $\gamma(0)$ | $N(E_F)/N_n$ |
|--------|-------------|-----------|-------------|--------------|
| T1     | 6.95        | no        | 3.1         | $\sim 15\%$ |
| U1     | 6.95        | yes       | 2.1         | $\sim 10\%$ |
| "     | 6.97        | yes       | 1.9         | $\sim 10\%$ |
| T2     | 6.95        | no        | 2.8         | $\sim 15\%$ |
| U2     | 6.95        | yes       | 2           | $\sim 10\%$ |

5 Discussion

5.1 Comparison with Lines of Nodes

The quadratic term is predicted to disappear in a magnetic field, where the energy dependence of the density of states close to the nodes, $N(E, H = 0) \propto |E - E_F|$, is replaced by a finite $N(E_F, H)$. The $\alpha T^2$ term is comparable in magnitude to the systematic uncertainty in the addenda $T^3$ phonon specific heat, but is several times larger than the relatively small systematic uncertainty of the field dependence, $< 1\%$. The global fit is thus the only possible way to identify the quadratic term in these data sets. The slight positive slope of the zero-field data set in Figure 2 can be described either as a phonon term which is about 5\% larger in zero field, which is difficult to explain, or as an $\alpha T^2$ term.

From the slope of the density of states in a superconductor with lines of nodes it is possible to predict $\alpha \approx \gamma_n/T_c \approx 0.2$ mJ/molK$^3$ for YBCO within factors of order unity [8], in good agreement with the value $\alpha = 0.1$ mJ/molK$^3$ obtained from the global fits (Table 1) on samples U1 and T1.

For the field dependence of the density of states, Volovik predicts $N(E_F, H) = \kappa N_n \sqrt{H/H_c}$ [6], where $\kappa$ is of order 1 and is defined by the vortex lattice structure, by the slope of the gap near the gap node, and by possible structure in $N_n$. Taking $\gamma_n = 20$ mJ/molK$^2$ [3, 4] and $H_{c2,\perp} = 150$ T gives the prediction $\gamma_\perp(H) = A\sqrt{H}$ with $A = \kappa \ast 1.6$ mJ/molK$^2 T^{1/2}$, in good agreement with the value $A = 0.9$ mJ/molK$^2 T^{1/2}$ found for both samples U1 and T1.

Both of the above comparisons used the measured value of $\gamma_n$, the angular averaged normal state density of states. Because both the $\alpha T^2$ and $A\sqrt{HT}$
terms are a consequence of the nodes, these terms should be sensitive to the normal-state density of states close to the nodes. The above comparisons may indicate that the density of states close to the nodes is smaller than the angular average of the density of states, as indicated by photoemission measurements on BSCCO [15].

These predictions do not distinguish between different types of lines of nodes, such as d-wave or extended s-wave [16]. In principle, this interpretation could also apply to a gap function with no nodes, but with a very small minimum gap, $\Delta \ll k_BT \approx 0.5 \text{meV}$. In order to produce these results, such a gap function would need to have an unusual energy dependence of the density of states similar to that associated with lines of nodes, $N(E, H = 0) \propto |E - E_F|$.

A quadratic $T^2$ term has been known for some time in the specific heat of heavy-fermion superconductors, and has recently been reported in the specific heat of La$_{2-x}$Sr$_x$CuO$_4$ [17]. Two other recent works support the existence of a $\sqrt{H}$ term in superconductors with lines of nodes: a reanalysis of existing data on polycrystalline YBCO samples shows a nonlinear $\gamma(H)T$ term which appears consistent with $\sqrt{H}$ [18], with similar but less dramatic curvature on Ca- and Sr-doped La$_2$CuO$_4$ samples, and a $\sqrt{H}$ term has also been recently reported in the low-temperature specific heat of UPt$_3$ [19].

5.2 Residual Density of States

The total residual density of states $N(E_F)$ in the superconducting state, as determined from the $\gamma(0)T$ term in the specific heat, is about 15% of the normal-state density of states for the twinned samples and about 10% for the untwinned samples (Table 2). The residual density of states can also be extracted from fitting the temperature dependence of the penetration depth [2] to the d-wave model with scattering of Hirschfeld et al. [5]. In samples similar to the samples measured here, these fits are inconsistent with a density of states which is greater than 1% of $N_n$ [3]. It appears difficult to reconcile the zero-field linear term $\gamma(0)T$ with the $\alpha T^2$ and $A\sqrt{HT}$ terms, and with the interpretation of the temperature dependence of the penetration depth. It is possible that the zero-field linear term has a separate origin from the excitations which give rise to the quadratic term and the field dependence of the specific heat. This assumption leads to a self-consistent analysis of the specific heat, and is given additional support by the measurements on the untwinned crystal U1, in which $\gamma(0)$ is decreased substantially but the $\alpha T^2$
and $A\sqrt{HT}$ terms are unchanged within statistical error bars.

### 5.3 Interpretation of the $A\sqrt{HT}$ term

Conventional high-$\kappa$ superconductors are expected to have a linear-$T$ term in the specific heat which is proportional to the volume of normal material in the cores, $c \approx \gamma_n TH / H_{c2}$, and such an $HT$ term has been observed in A15 superconductors. The analysis which predicts this $HT$ term does not apply to cuprate superconductors for two reasons. First, the small coherence length of the cuprates may result in a large excitation gap for quasiparticles which are confined to the vortex cores. Secondly, Caroli, deGennes, and Matricon assumed a fully gapped superconductor in showing that the quasiparticle excitations are confined to the vortex core.

Some form of a linear-$H$ term might be expected in the specific heat for any excitations which are confined to the vortex cores. Although there may in principle be some dependence of the core excitations on magnetic field, it is likely that for $H \ll H_{c2}$ the core excitations would be roughly independent of field. Thus, any signal resulting from these excitations would simply be proportional to the number of cores, or linear in $H$. The nonlinear field dependence of the observed $N(E_F, H)$ suggests excitations outside the vortex core.

The physical origin of the density of states predicted by Volovik is the same as the physical origin of the depairing current in a conventional superconductor. In the presence of a supercurrent with velocity $\vec{v}_s$, the quasiparticle excitation spectrum $\xi(\vec{k})$ is shifted by an amount $\vec{k} \cdot \vec{v}_s$. Far from the vortex core in a fully gapped superconductor, this shift is not large enough to change the density of states at the Fermi level, which is given by

$$N(E_F) = \frac{1}{(2\pi)^3} \int d^3k \int d^2r \delta \left( \xi(\vec{k}, \vec{r}) - \vec{k} \cdot \vec{v}_s \right).$$

(4)

For a superconductor with lines of nodes, the shift is significant everywhere that the superfluid velocity is not zero. Assuming a vortex superfluid flow $v_s \propto 1/r$, the local density of states at the Fermi level also falls off as $1/r$ (Figure 3). Integrating over an entire vortex, with the intervortex spacing $R(H)$ as the upper cutoff in the integral, gives a density of states per vortex $N(E_F, H) \propto R(H) \propto 1/\sqrt{H}$. Multiplying by the number of vortices, which is proportional to $H$, then gives the total density of states $N(E_F, H) \propto \sqrt{H}$.
The shift in the excitation spectrum depends on the angle between the local superfluid velocity and the nodes, giving the vortex the same symmetry as the gap (Figure 5). Because the nodes are 90° apart, the superfluid velocity can never be perpendicular to all nodes, and there are no places where $N(E_F)$ becomes zero. Both the density of states, and in a clean system the anisotropy in the density of states, extend throughout the entire vortex.

B. Parks et al. [23] have pointed out that their measurements of vortex dynamics in YBCO thin films are best explained by the presence of the extra quasiparticles expected for a vortex with an anisotropic gap. Recent theoretical work suggests that the structure of the vortex core in a d-wave superconductor is both interesting and complicated [6, 24, 25]. We have deliberately omitted the core region from Figure 5, since we are not aware of any model for the core which suggests that our measurements are probing the core states.

5.4 Predicted Parallel Field Dependence

Because $N(E_F, H)$ depends on the orientation of the current with respect to the nodes, improved measurements of the full angular dependence $\gamma_{\theta, \varphi}(H)T$ should be sensitive to the positions of the nodes. Define the orientation of the applied field $\vec{H}$ as $(\theta, \varphi)$, where $\theta$ is the polar angle measured with respect to the c-axis and $\varphi$ is the azimuthal angle measured with respect to the (110) crystalline axes. For tetragonal $d_{x^2−y^2}$ symmetry with antinodes along the Cu-O bonds, $\varphi = 0$ corresponds to a field parallel to a node (Figure 6). In this orientation, $(\theta = \pi/2, \varphi = 0)$, the currents in the ab-plane are flowing parallel to a single node. The density of states at the Fermi level is $\sqrt{2}$ larger at $(\theta = \pi/2, \varphi = \pi/4)$, where the currents flow parallel to the antinodes (Figure 6): in this orientation, the density of states picks up contributions from all of the nodes. The complete in-plane angular magnetic field dependence of the density of states is given by

$$N(0, H, \theta = \pi/2, \varphi) = \frac{\kappa I}{2} (|\sin \varphi| + |\cos \varphi|) N_F \sqrt{\frac{H}{H_{c2||}}}, \quad (5)$$

as shown by Volovik [26]. Using $\gamma_n = 20 \text{ mJ/molK}^2$ and taking $H/H_{c2||} = 0.005$ as an experimentally accessible field, the field-dependent linear term $\gamma_{||}(H)$ will vary from 0.7 to 1.0 mJ/molK$^2$ as a function of the azimuthal angle $\varphi$. This predicted angular dependence of the linear term in the specific
heat may be within achievable experimental resolution, although preliminary measurements at 8 Tesla on a heavily twinned sample with the field parallel to the twins and at a 45 deg angle to the twins did not detect any angular variation in $\gamma_\parallel(H)$.

6 Summary

The residual density of states at the Fermi level in the superconducting state of single-crystal YBa$_2$Cu$_3$O$_{7-\delta}$, $N(E_F, H = 0)/N_n$, is determined by the zero-field linear term $\gamma(H = 0)T$ in the specific heat. $N(E_F, H = 0)/N_n$ is qualitatively larger than would be expected from fitting the temperature dependence of the penetration depth [2] to expressions for lines of nodes with scattering [3]. In two twinned single crystals, $\gamma(H = 0)/\gamma_n \approx 0.15$, while in two untwinned single crystals, $\gamma(H = 0)/\gamma_n \approx 0.10$.

The specific heat also includes a $\gamma_\perp(H)T$ term, which obeys $\gamma_\perp(H) \approx \gamma_n \sqrt{H/H_{c2}}$ as predicted for superconductors with lines of nodes in the gap function [4]. This nonlinear field dependence of the density of states suggests quasiparticle excitations outside the vortex core, and appears to be independent of twinning, unlike the zero-field linear term. The in-plane angular magnetic field dependence of the specific heat, which is smaller than the perpendicular magnetic field dependence, is predicted to be sensitive to the locations of the nodes.

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Figure 1: Total specific heat at 0 and 8 Tesla for sample T1

Figure 2: Nonphonon specific heat for samples T1 and U1, with the global fit described in the text.

Figure 3: Coefficient of the linear term for crystals T1 and U1, determined by the global fit described in the text.

Figure 4: Coefficient of the linear term for crystal T1, determined by a) independent fits in which all parameters might be field-dependent, and b) a global fit with the $H = 0$ data set excluded.

Figure 5: Schematic of the local density of states $N(E_F)/N_n$ throughout a d-wave vortex.

Figure 6: The azimuthal field angle $\varphi$ relative to a tetragonal d-wave order parameter, and sketch of the current flows associated with an in-plane vortex for $\varphi = 0$ and $\varphi = \pi/4$. 
\( \gamma(0) + \gamma_{\perp}(H) \) (mJ/molK^2)
